Author Comment on „Space based observation of volcanic iodine monoxide“ by A. Schönhardt et al. (acp-2016-619)

Referring to the Interactive Comment of Referee #1, from 16 August 2016.

We are grateful for the comments, corrections and suggestions of Referee #1.

In the following, we address all the comments. Original comments are shown in black italics, our answers in black normal font and new text for the manuscript in blue.
All page and line references refer to the original ACPD online version, i.e. passages of changed text or locations for added text. Surely, the page/line numbers are different after revision.

1. Major Comments

Comment 1.1
When assessing the implications of detected iodine mass on stratospheric ozone, then I strongly recommend to consider the following scenarios for possible IO mixing ratios

- *a.) first a lower limit given by the largest amount of the air mass into which iodinated air is eventually mixed into, i.e. by assuming the emitted iodine is uniformly mixed into whole the atmosphere located above the maximum emission height (~150 mbar).*
- *b.) second an intermediate scenario an upper limit (the area of plume and layer of 4 km thickness), just in way as presented in the manuscript.*
- *c.) third an upper limit, where all iodine is mixed into lowermost stratosphere with the upper altitude located at 12 km, and a lower altitude given by the tropopause*

If you chose option b.) and/or c.) then the correct notion for the implications for ozone are that effects of volcanic iodine are 1.) regional, and restricted to the 2.) (subtropical?), midlatitude and high latitude UT/LS.

Answer to Comment
A global effect of IO can only be estimated if the emitted IO is spread over the entire atmosphere. In the manuscript we concentrate on the situation shortly after the volcanic eruption, therefore option b.) is our choice. We agree with the reviewer’s comment that the implications for ozone derived from the observed amounts of iodine are regional - and in addition also temporally restricted. The impacted region may be, however, dislocated from the volcano as the volcanic plume is transported over considerable distances within a short time of a few days.
After mixing and dilution of the emitted iodine, the volume mixing ratio (vmr) will be much smaller. Assessing a more global effect of the emitted iodine from the single volcanic eruption under consideration, the main influence is the large horizontal area over which the iodine is spread. The area of the volcanic plume is around $5x10^5$ km² in horizontal extent, while the entire globe has a surface area of around $5x10^8$ km², i.e. three orders of magnitude larger than the volcanic plume. For homogeneous mixing of iodine over the entire globe, the resulting vmr is a factor of 10^3 smaller than the derived 3 pptv within the volcanic plume a few days after the eruption. This yields a global averaged vmr of 0.003 pptv. Additional vertical mixing will take place which on the one hand further decreases the vmr, and on the other hand yields increasing vmr due to thinner air at higher altitude. All in all, the horizontal dilution is the main factor in this estimate.

Iodine chemistry in the atmosphere is likely to have a non-linear impact. For the revised manuscript we prefer to describe the regional and instantaneous impact, when considering the iodine within the volcanic plume. In addition we emphasize the fact that the implications for ozone are regional and in the first place affect the lower stratosphere or UTLS region. It is still important that the volcanic plume may move rather quickly so that the effects of iodine on the regional chemistry may be located at large distances from the erupting volcano.
Furthermore, we mention that for global estimates the horizontal dilution will enter with a factor of three orders of magnitude, so that on average the additional iodine vmr (resulting from the single

volcanic eruption under consideration) will be around 3 orders of magnitude smaller. The following paragraph is added to Section 4 in the revised version.

The above estimated IO vmr of 3pptv in the Kasatochi plume will be diluted with time. Spreading the released trace gas amount over the area of the entire globe decreases the vmr at the given altitude by three orders of magnitude as compared to the plume area. Consequently, strong implications for ozone depletion through iodine from a single volcanic eruption are probably mainly regional and restricted in time. Primarily, the lower stratosphere or UTLS region is affected. However, the region impacted by the emitted iodine may be dislocated from the erupting volcano due to the quickly moving volcanic plume covering distances of typically around several hundred km per day.

In addition, we change the wording in the summary adding the restriction of a „regional" reduction of ozone concentrations (p.12. ll. 19-20), cf. suggestions made by the reviewer in Comment 3.6.

Iodine volume mixing ratios of around 3 pptv may have substantial impact on the atmospheric composition, e.g., through regionally reducing the ozone concentrations.

Comment 1.2

Without further explanation (including chemical modeling), it is however unclear how the authors conclude from measured BrO, and IO vertical column amounts to the volcanic emission ratio of both halogens.

Answer to Comment

Certainly, the ratio of IO/BrO is not necessarily equal to the I/Br ratio. To our knowledge, the chemistry of iodine oxides within volcanic plumes is not yet implemented in chemical modelling systems. However, after the presented observations of large amounts of IO within the Kasatochi eruption plume, chemical modelling studies of volcanic iodine oxides are strongly encouraged.

The manuscript therefore concentrates on the observations and numbers of IO molecules. Nevertheless it is an interesting fact, that similar to other studies showing typically a one order of magnitude difference between bromine and iodine species in volcanic material (sediments, gas filter samples etc.), our measurements yield a one order of magnitude difference between gaseous BrO and IO in the emission plume.

Assuming that chemical conversions from halogens to halogen oxides are on a similar order of magnitude for iodine and bromine, then our satellite observations agree with these previous studies. While the observation of IO column amounts of only one order of magnitude smaller than BrO column amounts is an interesting fact by itself, the comment about the I/Br ratio is rather an additional thought. As stated in our manuscript, a detailed conclusion about how much iodine is emitted from Kasatochi certainly cannot be drawn from IO measurements alone without model calculations. However, the development of volcanic iodine chemistry for modelling purposes is out of scope of the present manuscript which focuses on observations. The above considerations and restrictions are therefore made more obvious and explicit in the revised manuscript.

(p.8, ll. 11-13)

Consequently, the emitted mass of iodine (3.9 to 10.8 t) can be regarded as a lower limit for the iodine content in the Kasatochi emission plume because this range is derived directly from the IO observations. Detailed chemical modelling would be needed to derive the total amount of reactive iodine in the volcanic plume from the observed IO column amounts by taking into account the other iodine species and all known chemical reactions that are taking place in the hot exhaust of the individual volcano. Such a modelling exercise is however out of scope of the current study.

(p.9, l.32)

Modelling of the halogen chemistry within the volcanic plume would be required to calculate the iodine and bromine amounts from the IO and BrO column observations. These estimates are performed under the given restrictions, and the present observations of volcanic emissions of IO shall encourage including chemistry of iodine and iodine oxides in volcanic halogen chemistry modelling in the future.

Also in the summary, one sentence has been changed, to remind readers of the restriction that only IO and BrO are measured but not the total iodine and bromine content in the plume.
(p.12 ll. 7-8)
Judging from the IO and BrO column amounts alone, this indicates a three order of magnitude difference with respect to the seawater ratio between iodine and bromine in agreement with previous filter measurements of volcanic samples at arc volcanos.

2. Minor Comments

Comment 2.1
I largely doubt that recent paper of Saiz-Lopez et al. (2015a) provide any cogent reference for stratospheric iodine, because

- *a.) Their definition of the TTL (see Figure 2, lower boundary 12 km upper boundary ~16 km) largely departs from the one given by Fueglistaler et al., 2008 (lower boundary 150hPa/355 K/14km, upper boundary 70hPa/425K/18.5km). A correct definition is very important in this context, since only air masses reaching altitude above the level of zero radiative heating (about 14.5 km, i.e. at the lower boundary of the Fueglistaler's TTL definition) may eventually enter the stratosphere.*
- *b.) the reported aircraft measurement of IO did not vertically extend into Fueglistaler's definition of TTL. Nevertheless they nicely complement previous iodine oxide measurements previously performed in the stratosphere (e.g., Pundt et al., 1998, Bösch et al., 2003, and 1998, Butz et al., 2009) to the upper troposphere.*
- *c.) the modelling in the study is at least in one aspect flawed, i.e. to correctly model the photolysis frequency and hence the ratio of IO/I_y at larger SZAs i.e., for the solar illumination when the balloon (solar occultation!) observations were made (inspect Figure S4, http://onlinelibrary.wiley.com/doi/10.1002/2015GL064796/full)*
- *d.) it missed to mention the study of Murphy et al., (2000) which claimed that iodine ions were found in the analyzed stratospheric aerosol. Evidently, if true the role 'reactive' iodine for stratospheric ozone may even further reduced.*

Accordingly I recommend you change the sentence (first page, bottom page) from….
Recent measurements and modeling studies conclude that iodine injection into the stratosphere is currently underestimated (Saiz-Lopez et al., 2015a), and that stratospheric amounts of reactive iodine lie between 0.25-0.7 parts per trillion by volume (pptv). ..to .. A recent study of most Saiz-Lopez et al., 2015a most optimistically estimate that stratospheric iodine may range between 0.25-0.7 parts per trillion by volume (pptv).
Here you also need to skip the notion reactive from the sentence, because the reported measurements detected a good deal of the total reactive iodine at daytime, i.e. (citation from the papers) [IO] ~ 0.17 ppt (at 12.5 km) on the aircraft (Dix et al., 2013, Saiz-Lopez et al., 2015a) and in Butz et al., (2009) …. 0.09 to 0.16 (+0.10/−0.04) ppt in the tropical lower stratosphere (21.0 km to 16.5 km) and 0.17 to 0.35 (+0.20/−0.08) ppt in the tropical upper troposphere (16.5 km to 13.5 km).

Answer to Comment
The evaluation of the reviewer regarding the study of Saiz-Lopez et al. (2015a) is understood. The addressed sentence (p.1, ll.25-26) in our manuscript will be changed in the revised version. In addition, the upper limits and observed values of the IO volume mixing ratio and estimates of total gaseous iodine derived by Pundt et al. (1998), Butz et al. (2009), as well as Volkamer et al. (2015) are now explicitly referred to.

From balloon borne observations, an upper limit for stratospheric iodine monoxide, IO, of 0.1 parts per trillion by volume (pptv) was determined in the tropics (Butz et al., 2009), while upper limits for IO of 0.2 pptv at 20 km, or 0.1 pptv at 15 km (Pundt et al., 1998) were derived in the mid and high latitudes. Butz et al. (2009) conclude upper limits of total gaseous iodine of about 0.09 to 0.16 pptv in the tropical lower stratosphere (21.0 km to 16.5 km) and 0.17 to 0.35 pptv in the tropical upper troposphere (16.5 km to 13.5 km). A recent study by Saiz-Lopez et al. (2015a) estimates that stratospheric iodine may range between 0.25-0.7 pptv. This is based on, e.g., new aircraft observations in the tropics from which volume mixing ratios of IO between 0.1–0.2 pptv at altitudes up to 14 km were obtained (Volkamer et al., 2015).

Comment 2.2

Further the authors need to provide a reference to the following sentence: Even at sub-pptv levels, reactive iodine may significantly impact on stratospheric ozone chemistry….. c.f., Hossaini et al., 2015 (inspect Figure 2a), WMO (2014) and references therein ... and considering them probably it is worthwhile to rethink the notion significantly in the sentence.

Answer to Comment

Effects of iodine vmr of 1pptv as well as at sub-pptv level have, e.g., been analysed by Solomon et al., 1994. Although some reaction rates were not known at that time, conclusions are that stratospheric iodine may play a role in ozone depletion. Also Hossaini et al., 2015, find an influence of very-short lived iodine species on stratospheric ozone levels when considering the small amount of 0.15 pptv of iodine. Although the overall importance of iodine is less that of bromine, which has much larger stratospheric abundances, the iodine effect on ozone is still noticeable. As suggested, the notion "significantly" will be omitted from the respective sentence in our current manuscript. In addition, references to (Solomon et al., 1994) and (Hossaini et al., 2015) are added.

(p.2, ll. 1-2)

Even at sub-pptv levels, reactive iodine may impact on stratospheric ozone chemistry (Solomon et al., 1994; Hossaini et al., 2015).

3. Recommended editorial changes and typos

The manuscript contains are larger number of oddities with English, proof-reading of the manuscript by a native English speaker is highly recommended.

Comment 3.1

Examples of oddities with the grammar/style/notation (a selection)
3.1 on page 21 (and at many other places): The GOME-2A data show higher noise levels than the respective SCIAMACHY measurements. Consequently, for the analysis of GOME-2A data, the use of more spectral information from a larger fitting window was (is!!) investigated. So please consider the coherence in the grammatical tense i.e., I largely recommend that you use the simple presence in all sentences we you do not explicitly refer to past, or future event, and that something started in the past and is still ongoing et cetera….
>> Done. (page 4)

More example:

- *Page 4 line 24: ….was investigated → is investigated*

>> Done.

- *Page 4 line 24: For SCIAMACHY, the 3T retrieval had not been successful (Schönhardt et al., 2008) → For SCIAMACHY, the 3T retrieval was not successful (Schönhardt et al., 2008)*

>> Done.

- *Page 4 line 30: For all IO retrievals, a daily averaged Earthshine spectrum was used as reference background → For all IO retrievals, a daily averaged Earthshine spectrum is used as reference background.*

>> Done.

- *Page 7, line 21: For IO, values in that region were lower so the effect is less pronounced. .... IO vertical column amounts are lower in this region, accordingly this effect (and please name the effect it properly!) less pronounced.*

>> The sentence is changed to:
For IO, values in that region are lower. Consequently, the influence of the precise plume shape on the calculation of the integrated amount is less pronounced.

- *Page 8, line 24: Previous satellite studies have observed that → Previous satellite studies reported that often BrO……*

>> Done.

- *Page 30, line 30: The IO and BrO data from within a rectangular latitude-longitude box enclosing the entire volcanic plume has been investigated individually for each day between August 08 and 12…-> The IO and BrO vertical column amounts enclosing the entire volcanic plume with rectangular in latitude-longitude is investigated for each individual day between August 08 and 12, 2008.*

>> (Page 8) The sentence now reads:
The IO and BrO vertical column amounts that are observed within a rectangular latitude-longitude box which encloses the entire volcanic plume are investigated for each individual day between August 8 and 12, 2008.

- *Page 10, line: Hydrothermal experiments have been used to analyse the compositions of hydrous fluids and silicate melts with respect to the different halogens (Bureau et al., 2000) ....-> Results of hydrothermal experiments were used to analyse the compositions of hydrous fluids and silicate melts with respect to the different halogens (Bureau et al., 2000). (were.... Since Bureau et al., 2000 used it once in the past)*

>> Done.

- *Page 10, line 26: For several days following the major eruption of Kasatochi volcano in August 2008, iodine monoxide was observed by satellite → Following the major eruption of the Kasatochi volcano in August 2008, iodine monoxide is observed by satellite in the volcanic plumes for several days.*

>> Done.

Comment 3.2
3.2 Wrong units and notation
Example:
On line page, line 10: The columns of IO are approximately....instead of .... The columns amounts of IO..... because IO does not tend to form columns, but a certain number of IO molecules reside in an air column, and (2) if you name and/or define thing in a manuscript, please keep the same name and/or definition throughout the manuscript in order not to confuse the reader.
>> Done (Page 1). Only vice versa:
The sentence now reads "The column amounts of IO…" instead of " The columns of IO…"

More examples:

- *Page 4, line 19: The comparably large IO columns are connected -→ The comparably large IO column amounts are detected with*

>> Done.

- *Page 5, line 10: The DOAS analysis yields the trace gas slant column values→ The DOAS analysis yields (differential) slant column amounts…*

>> Done.

- *Page 5, line 10: In order to convert these numbers ..... → In order to convert the slant column amounts.....*

>> Done.

- *Page 6, line 29: 3.3 Analysis of IO and BrO amounts → 3.3 Analysis of IO and BrO column amounts*

>> We prefer to keep the title of the section because the analysis deals with the integrated IO and BrO amounts in addition to and derived from the observed IO and BrO column amounts.

- *Page 7, line 27: The range of IO values between 4.3 and 12.1 t .....-> The integrated mass of IO ranges between 4.3 and 12.1 t*

>> See next comment.

- *Page 7, line 27: The range of IO values between 4.3 and 12.1 t corresponds to an amount of reactive iodine between 3.9 and 10.8. -> The IO mass ranges between 4.3 and 12.1 t, which corresponds to mass of reactive iodine between 3.9 and 10.8t. And!!! how are the ranges calculated? Here any reader becomes confused because a result is reported before it is outlined how the result is obtained. So there is also problem with the logical order.*

>> The method how the integrated amount of IO molecules is explained above the results given in the respective sentence. The conversion from integrated mass of IO to integrated mass of iodine refers to the iodine directly contained in the IO, and the calculation uses the molar masses of iodine and oxygen. The addressed sentence is changed to:
The integrated mass of IO hence ranges between 4.3 and 12.2 t. Using the molar masses of iodine and oxygen, this amount of IO contains an integrated mass of reactive iodine between 3.9 and 10.8 t.

- *Page 8, line 3: On August 12 an amount of 66 t of BrO remains…-> On August 12, 2008, an integrated mass of 66 t of BrO remains in the atmosphere.*

>> In order to avoid misunderstandings (as there is additional BrO in the atmosphere apart from the integrated mass of BrO in the volcanic plume), the sentence is changed to:
On August 12, 2008, an integrated mass of 66 t of BrO remains in the volcanic plume.

In addition, the sentences before and after the above addressed sentences are also changed in analogy to the analyses of the integrated mass of IO and iodine:
The integrated mass of BrO within the plume increases from 26 t on August 8 to 76 t and 79 t on August 9 and 10, and reaches a maximum of 87 t on August 11.
Directly converting the integrated mass of BrO between 26 and 87 t to the corresponding integrated mass of reactive Br, a range between 22 and 73 t is derived, using the molar masses of bromine and oxygen.

- *Page 8, line 21: ... retrieved amounts of … instead of …retrieved column amounts ….*

>> Vice versa. Wording is changed from "amounts" to "column amounts" in this sentence, and also in the next sentence, as well as three and five sentences further down.

Comment 3.3

3.3 A wrong comparative!

>> Probably the reviewer refers to the second sentence of section 3.3. The sentence has been changed from: … IO retrieved from GOME-2A yields a more accurate analysis of the total iodine amount and mass emitted from the Kasatochi eruption.
to: … GOME-2A IO results provide a more accurate analysis of the total iodine amount and mass emitted from the Kasatochi eruption than the IO results retrieved from SCIAMACHY.

More examples

- *Page 6, line 15: Due to the much better spatial coverage of the GOME-2A instrument, the IO plume….. Due to the much better spatial coverage of the GOME-2A instrument as compared to (the SCIAMACHY?) instrument, the IO plume…*

>> Done.

- *Page 10, line 1: A wrong comparative! … the ratio of Cl vs. I is about two orders of magnitude lower than in seawater. … the ratio of Cl vs. I is about two orders of magnitudes lower in volcanic plumes than in seawater.*

>> Done.

- In addition, we have changed the sentence on p.8, l.25:

>> For the two depicted cases, the IO column amount is also lower in the plume center than in some areas around the center.

Comment 3.4

3.4 Please provide appropriate dates in order to improve the readability of the manuscript: For example on page 7 lines, 15, 18, 19 …. i.e. August 8 to 11, 2008 instead of August 08 to 11, or August 12, 2008 instead of August 12, …

>> The day format has been changed from, e.g., August 08 to August 8, throughout the manuscript. In addition, the year 2008 has been added in most places. It has not been added to all dates though, e.g., if the range August 8 to 12, 2008, is given first and then several single dates from this range are mentioned individually. Adding the year in all of those places would again degrade readability.

Comment 3.5

3.5 Page 12, lines 9 and 10: Iodine shows a stronger preference than bromine to partition into volcanic fluids than volcanic melts in the volcanic chamber below the volcano.-> Iodine shows a stronger preference than bromine to partition into volcanic fluid than melt in the volcanic chamber located underneath the volcano. (it is necessary to erase the second volcanic before melt, otherwise the sentence is ambiguous).

>> Done.

Comment 3.6

Recommended editorial changes and typos (a selection)

- *Page 2, line 12: In the Polar troposphere, both, bromine and iodine oxides are observed ……..*
  → In the polar troposphere, bromine and iodine oxides are both observed

>> Done.

- *Page 4, line 31: ……………which is assumed to have small column amounts of IO..-> which is likely to contain small column amounts of IO*

>> Done.

- *Page 5: The header of Table 1 need to appear on top of the table.*

>> Done. The caption of Table 2 is also moved to the top of the table for the revised version.

- *Page 5, line 13: This is an adequate assumption for the current study as the volcanic plume is located at fairly high altitudes (Theys et al., 2009) and the relevant SZA values are below 50°. → For the current study, assuming a geometric AMF is adequate since the volcanic plume is located at fairly high altitudes (Theys et al., 2009) and the relevant SZA < 50°.*

>> Sentence changed to:
For the current study, assuming a geometric AMF is adequate since the volcanic plume is located at fairly high altitudes (Theys et al., 2009) and the relevant solar zenith angle is below 50°.

- *Page 6 line 19: The results including both GOME-2A retrievals as discussed in Sec. 2.2 are summarized in Tab. 2 and the corresponding spectral fits are shown in Fig. 3.-> .... The IO and BrO retrieval for the GOME-2A instrument is discussed in Sec. 2.2 and the results are summarized in Tab. 2. The corresponding spectral fits are shown in Fig. 3.*

>> Table 2 deals with the different IO retrievals, one from SCIAMACHY and two from GOME-2A, but not with the BrO retrieval. Therefore, the suggested changes for the addressed sentence are not appropriate. Instead, the sentence is changed to:
The comparison includes the IO from the SCIAMACHY retrieval as well as IO from both GOME-2A retrievals, and the results are summarized in Tab. 2. The IO retrieval settings are discussed in Sec. 2.2 and the corresponding spectral fits are shown in Fig. 3.

- *Page 5, line 20: Following the eruption of Kasatochi, enhanced IO amounts are visible for several days. → Post the eruption of Kasatochi, enhanced IO column amounts are detected within the plumes for several days.*

>> Accepted with slight changes in the word choice:
After the eruption of Kasatochi, enhanced IO column amounts are detected within the volcanic plume for several days.

- *Page 6 line 21: GOME-2A results also show good retrieval quality with a relative retrieval error of around 14 %, which is somewhat larger than for the SCIAMACHY examples.-> The GOME-2A spectral retrieval are also of good quality with relative retrieval errors of around 14 %. The retrieval errors is thus larger than for the retrieval of SCIAMACHY data.*

>> Accepted with slight changes, especially in the grammar:
The GOME-2A spectral retrievals are also of good quality with relative retrieval errors of around 14 %. The retrieval error is thus larger than for the SCIAMACHY retrieval.

- *Page 6, line 24: .....depending on several factors such as light intensity……. What is a light intensity. Is it a radiance 8yes) or irradiance. Accordingly you need to change the sentence to: … .....depending on several factors, such as the received skylight radiance and …*

>> Accepted, while omitting the word skylight.

- *Page 6, line 25: For the example case, GOME-2A detects slightly less IO than SCIAMACHY, however, in other collocation cases the relation is reverse.-> For the discussed (shown) examples, the GOME-2A instruments detects slightly less IO than the SCIAMACHY instrument. On other collocation events the relation is however reversed.*

>> Done.

- *Page 6, line 27: For a rapidly moving volcanic plume, in which relatively fast and complex multiphase photochemical reactions take place, some real differences in the IO amounts as seen by the two instruments are therefore expected. → For rapidly moving volcanic plumes, differences in the detected IO column amounts by the two instruments are expected, either as a matter of changing IO concentrations due to relatively fast and complex multiphase photochemical reactions, the size of the ground scene and or changing ground or cloud albedo.*

>> Done.

- *Page 6, line 30: The sampling of GOME-2A measurements is intrinsically higher than that of SCIAMACHY → The sampling of spectra by the GOME-2A instrument is intrinsically better than that of the SCIAMACHY instrument*

>> Done.

- *Page 7, lines 1 and 2: …where xx is the mean IO vertical column and xx is the standard deviation, both derived from measurements on the days before the eruption …. ->…where xx is the mean IO vertical column and xx is its standard deviation. Both parameters are derived from measurements on the days before the eruption ….*

>> Done.

- *Page 7, line 6: ……….are calculated using the data from three days with satellite overpasses before ……….are calculated using the data from three consecutive days of satellite coverage prior…*

>> Done.

- *Page 7, line 22: ….. but due to the latter observation ….-> … but due to the latter finding*

>> Done.

- *Page 8, line 13: Consequently, the amount of iodine between 3.9 to 10.8 t derived here from the IO alone needs to be considered as a lower limit.-> Consequently, the emitted mass of iodine (3.9 to 10.8 t) can be regarded as a lower limit.*

>> Done. The sentence is changed as part of the answer to Comment 1.2 and now reads:
Consequently, the emitted mass of iodine (3.9 to 10.8 t) can be regarded as a lower limit for the iodine content in the Kasatochi emission plume because this range is derived directly from the IO observations.

- *Page 8, line 113: The amount of iodine derived from the Kasatochi eruption is of the same order of magnitude determined by measurements at degassing volcanos for one 15 year, e.g. 10 t/yr of iodine at Mt. Etna, Italy, (Aiuppa et al., 2005) or 12 t/yr at Satsuma-Iwojima…-> The emitted mass of iodine inferred for the Kasatochi eruption on …. is of the same order of magnitude as previously determined for the annually integrated flux for degassing volcanos, e.g. 10 t/yr of iodine at Mt. Etna, Italy, (Aiuppa et al., 2005) or 12 t/yr at Satsuma-Iwojima…*

>> Done.

- *Page 9, line 9: As a consequence, degassing from the magma may take place at different pressure, i.e. at different depth and time, for the two halogen species. -> For the two halogen species degassing from the magma may take place at different pressures, i.e. at different depth of the volcanic abyss (erase …and time).*

>> Done.

- *Page 9 line 1 to 16: Move to the discussion section.*

>> The content of page 9, lines 1 to 9 and lines 13 to 16, is moved to the discussion section. The content of lines 10 to 13, however, describes immediate observations and the comparison of IO, BrO and SO_2 spatial distributions. This part needs to remain in the current section. Linking sentences have been included along with the two moved parts.

The following sentence has been added prior to the first moved part in the discussion section:
The spatial distributions of IO, BrO and SO_2 are described in Sec.3.4, and some differences between the three species are observed.

The following sentence has been added prior to the second moved part in the discussion section:
In addition, some clear differences between the spatial distributions of the halogen oxides and SO_2 are found.

After deleting the mentioned passages, the following closing remark is now given at the end of Sec. 3.4.
The three different trace gases observed by satellite hence show several individual aspects in their spatial distribution within the volcanic plume.

- *Page 9, line 21: The corresponding ratio for the mass of BrO to IO lies between 4.0 and 6.7 (2.8 on day August 08), using data from Fig. 4. _> The corresponding mass ratio for BrO to IO range between 4.0 and 6.7, and 2.8 on August 08, 2008…. A question: Do you refer to a mass or a number density ratio?*

>> As it says in the manuscript, the values in the addressed sentence refer to a mass ratio. (The numbers given in the sentence prior to the addressed sentence refer to the number density: "For the individual days August 9 to 12, 2008, the ratio for the integrated number of BrO to IO molecules lies between 6.7 and 10.0 (4.2 on August 08).")
The two sentences are changed to:
For the individual days from August 9 to 12, 2008, the ratio for the integrated number of BrO to IO molecules lies between 6.7 and 10.0, and amounts to 4.2 on August 8, 2008. The corresponding mass ratio for BrO to IO ranges between 4.0 and 6.7, and amounts to 2.8 on August 8, 2008, using data from Fig. 4.

- *Page 9, line 23: Data from four days August 09 to 12…-> Data from the period August 09 to 12, 2008….*

>> Changed to:
Data from the four day period August 9 to 12, 2008, is included in the comparison.

- *Page 9, line 28: The uncertainties given by the latter study are rather large, but the results agree on the difference of one order of magnitude between the two halogens. → The uncertainties in emission fluxes given by the latter study are rather large, but the two halogens the results agree within their error bars.*

>> Sentence changed to:
The uncertainties in emission fluxes given by the latter study are rather large, but the results for the halogen flux ratios agree with the present satellite observations within their error bars.

- *Page 9, line 32: This implies that the iodine vs. bromine ratio in the volcanic plume is enhanced by about three orders of magnitude judging from the IO and BrO observations. -> By considering IO and BrO observations, the iodine vs. bromine ratio is enhanced by about three orders of magnitudes in volcanic plumes as compared to sea water.*

>> Sentence changed to:
By considering singly the IO and BrO observations, the iodine vs. bromine number ratio is thus enhanced by about three orders of magnitude in the volcanic plume as compared to sea water.

- *Page 10, line 13: The temporal evolution of the observed IO and BrO amounts in the plume is of interest.-> Of interest is also to study the temporal evolution of the observed IO and BrO column amounts within the plume.*

>> Done.

- *Page 10, line 18: Different chemical pathways and time constants for IO and BrO production and destruction will influence the temporal variation in the ratios. -> The different chemical pathways and time constants for IO and BrO production and destruction also influences the temporal variation of the I/Br ratio.*

>> Sentence changed to:
The different chemical pathways and time constants for IO and BrO production and destruction also influence the temporal variation of the I/Br ratio.

- *Page 10, line 26: The retrieved number of IO molecules of around 5×1028 molecules from days August 10 and 11…-> The retrieved total number of IO molecules of amounts about 5×10^{28} molecules for August 10 and 11, 2008….*

>> Sentence changed to:

The retrieved integrated number of IO molecules of about 5×10^{28} molecules for August 10 and 11, 2008, is used as lower limit of the emitted iodine amount.

- *Page 10, line 26: The lateral plume extent on these days lies around 5×10^5 km^2.. -> On both days the plumes extents over 5×10^5 km^2*

>> Sentence changed to:

On both days the plume extends horizontally over 5×10^5 km^2.

- *Page 10, line 30: An iodine abundance of 3 pptv strongly impacts, e.g., on the ozone levels..-> Iodine mixing ratios of 3 pptv may strongly impact ozone concentrations….*

>> Sentence changed to:

Iodine mixing ratios of 3 pptv may have a strong impact on ozone concentrations….

- *Page 10, line 32: The Kasatochi plume altitude reaches into the lower stratosphere. Consequently, the satellite observations of large amounts of iodine after the Kasatochi eruption indicate, that volcanic eruptions may lead to a substantial input of iodine to the stratosphere as well as the upper troposphere lower stratosphere (UTLS) region, depending on the eruption altitude and plume rise. → The upper part of the Kasatochi plume may have reached into the lower stratosphere. In consequence, our satellite-based observations of iodine oxide indicates that volcanic eruptions may have an impact on the iodine concentrations at least regionally in the upper troposphere and lower stratosphere (UTLS).*

>> Done. Second sentence with minor modifications/corrections:

Consequently, the presented satellite-based observations of iodine monoxide indicate that volcanic eruptions may have an impact on the iodine concentrations at least regionally in the upper troposphere and lower stratosphere.

- *Page 10, line 2: In this case the lower stratosphere will be most affected -> In this case the lower stratosphere may become most affected.*

>> Done. (Page 11)

- *Comparing these numbers to the results of BrO, the ratio for BrO to IO molecules lies between 6.7 and 10.0…-> By rationing the masses of the two halogen oxides found within the plume BrO, then the BrO/IO ratio ranges between 6.7 and 10.0…*

>> This would change the original meaning of the sentence, which addresses the (number of) molecules and not the masses of IO and BrO. The sentence is clarified by:

By comparing the integrated numbers of IO and BrO molecules found within the volcanic plume, the BrO/IO number ratio ranges between 6.7 and 10.0, while the BrO/IO mass ratio lies between 4.0 and 6.7.

- *Page 12, line 20 An iodine vmr of around 3 pptv has substantial impact on the atmospheric composition, e.g., through reducing the ozone levels.-> Iodine volume mixing ratios of around 3 pptv may have substantial impact on the atmospheric composition, e.g., through regionally reducing the ozone concentrations.*

>> Done.

- *Page 12, line 22: ……………..and will be facilitated in the future…-> and in future will be facilitated*

>> Done.

4. Additional references

1. Butz, A. H. Bosch, C. Camy-Peyret, M. P. Chipperfield, M. Dorf, S. Kreycy, L. Kritten, C. Prados-Roman, J. Schwärzle, and K. Pfeilsticker, Constraints on inorganic gaseous iodine in the tropical upper troposphere and stratosphere inferred from balloon-borne solar occultation observations, Atmos. Chem. Phys., 9, 7229 – 7242, 2009.

2. Fueglistaler, S., A. E. Dessler, T. J. Dunkerton, I. Folkins, Q. Fu, and P. W. Mote, Tropical tropopause layer. Reviews of Geophysics, Vol. 47, No. 1, n/a–n/a, 2009.

3. Hossaini, R., M. P. Chipperfield, S. A. Montzka, A. Rap, S. Dhomse, and W. Feng, Efficiency of short-lived halogens at influencing climate through depletion of stratospheric ozone, Nature Geoscience, DOI: 10.1038/NGEO2363, Feb. 15, 2015.

4. Murphy, D. M. and Thomson, D. S.: Halogen ions and NO+ in the mass spectra of aerosols in the upper troposphere and lower stratosphere, Geophys. Res. Lett., 27, 3217–3220, doi:10.1029/1999GL011267, 2000.

5. Pundt, I., Pommereau, J., Phillips, C. et al. Upper Limit of Iodine Oxide in the Lower Stratosphere, Journal of Atmospheric Chemistry (1998) 30: 173. doi:10.1023/A:1006071612477

>> References 1, 2, 4 and 5 are used by the reviewer to support the evaluation of the study by Saiz-Lopez et al., 2015a. The individual points of criticism are not meant to be incorporated in the revised version of our manuscript. Reference 3 is used in Comment 2.2 as basis for the evaluation of the iodine impact on stratospheric ozone chemistry. The study by Hossaini et al. (2015) is therefore cited in the revised version as stated in the answer to Comment 2.2.
Due to their direct relation to the content of the present study, we additionally include Reference 1 and Reference 5 in the introduction, as well as Reference 4 in Sec. 4 of the revised version.

Reference 4 is also linked to a comment by Reviewer #2 about iodine forming aerosol particles. The following paragraph is included in the revised version.

The evolution of iodine species in the volcanic plume may be further affected by particle formation and heterogeneous reactions. Murphy and Thomson (2000) measured enhanced iodine content in aerosols in the upper troposphere and lower stratosphere (UTLS) region. This finding has two further implications. Particles may serve as a sink for iodine reducing the availability of reactive iodine, and on the other hand they may provide pathways for heterogeneous reactions from which reactive iodine compounds may be released again.

Further reference included in the revised version (introduction) in the context of the comments by Referee #1:

Volkamer, R., et al.: Aircraft measurements of BrO, IO, glyoxal, NO_2, H_2O, O_2-O_2 and aerosol extinction profiles in the tropics: Comparison with aircraft-/ship-based in situ and lidar measurements, Atmos. Meas. Tech., 8(5), 2121–2148, 2015.