Anonymous Referee #1

This manuscript analyses the variability in CHBr3, as one of the major VSLS Bromine species that can be transported to the stratosphere by strong convection in the tropical latitudes. They have used a representative surface emission pattern at monthly-mean time intervals, and transport-chemistry parameterized using FLEXPART, a widely used modelling system for short-lived species simulation. The manuscript is generally very well written, and the subject of this research is important. However, I have some strong reservations which have to be addressed before manuscript can be accepted for publication in ACP.

We thank Referee 1 for his/her valuable comments. We have changed the manuscript according to the comments listed below and think that these changes have improved the quality of the manuscript.

Major comments.

I am very concerned with the model simulation over the South Asia region. The pattern of transport of VSLS species from the surface layers appears unrealistic (Fig. 8), meaning the location of the anticyclone in JJA appears to be vastly misplaced for both CHBr3 and Transit time, based on our knowledge of global (Eulerian) chemistry-transport models and observation of tracers, e.g., Park et al. (JGR, 2007) or Chandra et al. (ACP, 2017). The upward motion over the India monsoon domain is limited to northern India rather than in the tropical areas.

The referee is correct that during boreal summer, the centre of the anticyclone is situated over the continental part of Asia extending from the Tibetan Plateau to the Middle East and north-eastern Africa. As various modelling and observational studies have demonstrated, intense convection can loft boundary layer pollutants and aerosols into the upper troposphere/lower stratosphere anticyclone. Here the polluted air masses remain mostly confined within the closed circulation of the anticyclone for days to weeks

When comparing this current state of knowledge with our results, it is important to keep in mind that 1) nearly all studies focus on pollutants emitted over the Asian land masses and not on oceanic trace gases and vice versa 2) air masses found in the anticyclone mostly originate from the planetary boundary and not from the marine boundary layer.

The second point is demonstrated by Bergmann et al. (2013) who state: ' … We calculated regional contributions to air within the anticyclone by boundary layer sources from five regions: the Tibetan Plateau, India/SE Asia, eastern China, the Indian Ocean, and the western Pacific. … Boundary layer sources for the anticyclone are primarily from the Tibetan Plateau and India/SE Asia (a combined 70%–80%) at both the 200 mbar and 100 mbar levels, with minor contributions by the western Pacific and Indian Ocean (a combined 15%–20%). …'

Furthermore Vogel et al. (2018) based on CLAMS backward simulations of the anticyclone states '… High fractions of air from India/China up to 90 % and low fractions below 10 % from the tropical adjacent regions are found in the core of the Asian monsoon anticyclone at 360 K potential temperature. Highest fractions from the tropical adjacent regions of about 40 % are found in a belt around the edge of the anticyclone …'

In consequence the stratospheric entrainment of an oceanic trace gas, like bromoform, over the South Asian region does not necessarily need to reflect the entrainment of anthropogenic pollutants like CO. As the air trapped in the anticyclone only contains a small fraction of marine boundary layer air, marine trace gases as short-lived as bromoform might not show a maximum in the anticyclone. Localized convection extending from the Arabian Sea to the Philippines including the Bay of Bengal and the Arabian Sea acting on a trace gas with large sources over the equatorial and inner tropical oceans can be expected to cause high entrainment over southern India and adjacent ocean. This is consistent with results presented in Fiehn et al. (2018), who used the same FLEXPART ERA-Interim model experiment set up as our study and found the main injection of bromoform during boreal summer over the southern tip of India associated with the high local oceanic sources and strong convection of the summer monsoon.

Which raises a wider question about the validity of the FLEXPART model framework implemented in the work. Can there be some independent tracer simulation to validate the model transport, say using Radon-222 or SF6 or CO for which we have better knowledge of model transport (Forster et al., 2007 could be using older version of the model, is your version same that work?). I am aware that the FLEXPART works reasonably well for the horizontal transport at regional scale but not sure whether suitable for such global model simulation.

FLEXPART has been successfully used in studies of transport and tracer distributions in the Asian monsoon (e.g., Lal et al., 2014; Lelieveld et al., 2018; Li et al., 2016). Some of these studies include comparisons to or evaluations of observational data. In previous related work, we have validated our VSLS FLEXPART ERA-Interim model set-up for the tropical West Pacific ship-aircraft-land campaign in November 2011 which showed very good agreement between modelled and observed VSLS profiles in this convection dominated area (Fuhlbruegge et al 2016 ACP; "The EU Project SHIVA" special issue ACP).
Validation of the convective scheme used in FLEXPART by Forster et al., 2007 is indeed based on an older version of FLEXPART compared to the one used in our paper. However, the convective scheme has not been changed in between versions, therefore the presented validation still applies to the current version of the model.

I am also very disappointed in the ways the model and measurements are compared. For a reasonable evaluation of the model simulations we need to first sample the model at the time and location of measurements.

We have improved our model-measurement comparisons by sampling the model output at the measurement time and location for comparisons in Figures 4 and 7. The new comparison shows slightly different results; however, the overall findings do not change substantially.

Minor comments.

p.3, l#4: could you put a timeline here, as in the WMO/SAOD, 2018
Timeline and reference have been added.

p.3, l#13: may be cite - Hossaini et al., ACP, 2016
Reference has been added.

p.9, l#1ff: this is quite a bit of simplification of the loss of SGs, a proper treatment using a 3D OH field would have been useful to simulate tropospheric distribution of the SGs.

In general, FLEXPART simulations can also be carried out with the chemical loss term derived from the reaction with OH based on a prescribed 3D OH field. The chemical loss of bromoform, however, is largely driven by photolysis (Hossaini et al., 2010, Figure 4), which is not included in the current FLEXPART version. Therefore, the chemical loss is based on a lifetime profile derived from detailed chemistry simulations.

p. 11: Figure 1 and associated text: the southern spread of this anticyclone as appears from your figure looks too wide, is there a possibility to show these plots for summer and winter (arrange a 2x2 panel figure). The panel b doesn't provide sufficient information. Also, I would like you to add the surface fluxes for the two seasons, making it 2colx3row figure. Even though the emissions are taken from Ziska et al., the readers cannot directly assess results without showing here again.

We have added seasonal composites of the $CHBr_3$ concentrations at 17 km and of the air-sea fluxes from Ziska et al. (2013) to the supplementary material.

Figure 3 and associated text: Observations are much less than the model values

This is true for the AVE campaign but not for Pre-AVE or for ACCENT (see also the summarized comparison in Figure 4).

How to explain the spotty features in the model simulations?

The spotty features in the model simulations are a result of the high oceanic sources directly underneath interacting with localized convective transport. The latter brings localized air masses with very high bromoform mixing ratios from the boundary layer into the 15-17km layer. We have added this explanation to the manuscript.

Observations are higher over the Gulf of Mexico persistently, hinting towards low emissions?

Not necessarily, as the observations are in very good agreement for the Pre-AVE campaign and even lower for the AVE campaign

Figure 4 and associated text: I am not very happy with this comparison. I believe that the model simulations should be first sampled at the measurement points and then compared with measured values.

We have improved the comparison by adding model values sampled at the time and location of the measurements. We decided to also keep the averaged model values in the Figure in order to highlight the seasonal cycle and high tempo-spatial variability of $CHBr_3$ in the upper TTL suggested by the model results.

This plot makes me interpret that the model simulation is out of phase with the measurements during June to Aug! I do not know if this problem comes from not sampling the model properly or the model itself is wrong.

It is true that during the NH summer months 3 out of 4 campaigns show lower values. However, the ACCENT campaign shows much higher values. Most important, the standard deviation of the model results suggest higher variability during this time which is confirmed

by the relatively large differences between the individual campaigns. Given the latter it is not clear if the model overestimates the NH summer entrainment. The manuscript states " … For the campaigns during NH summer, mean differences are larger in most cases. At the same time, the temporal and spatial variability of the simulated CHBr3 distribution is also larger so that nearly all observations fall within the simulated uncertainty. The large differences between the individual campaigns during NH summer confirm the increased variability suggested by the model results. …".

p.21, l18: Again, good to have shown the emission maps in Fig. 1, for winter and summer.

We have added seasonal composites of the air-sea fluxes from Ziska et al. (2013) to the supplementary material and added a reference to the text.

Figure 7 and associated text: This again reiterate why are you not showing one-to-one comparison, given that the transport varies

We have improved the comparison by adding model values sampled at the time and location of the measurements. The new comparison shows slightly different results; however, the overall findings do not change substantially.

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Anonymous Referee #2

The manuscript presents a study of the transport of CHBr3 and CH2Br2, concentrating on how this might have changed over the past decades. They use a now widely-used static global sea-surface concentration field for these species, then calculate monthly fluxes based using offline meteorology. This is run through a Lagrangian model using emissions on a monthly basis. This is an area of study that is of interest to a wide audience and is appropriate in scope for ACP.

We thank Referee 1 for his/her valuable comments. We have changed the manuscript according to the comments listed below and think that these changes have improved the quality of the manuscript.

I can understand that the approach described here could yield information about the source gas delivered to the TTL/UTLS, however, I am not convinced that it is an appropriate methodology for understanding product gases. A largely expanded discussion of uncertainty is needed, as well as comparisons with product gases or referencing of this work elsewhere. The comparisons with inorganic bromine constraints could provide evidence to the argument that this methodology can give valid insights into product gas delivery to the stratosphere. Clearer and more detailed explanations are needed for this approach, as these kinds of studies have more often used models with online chemistry with the online process (e.g. photolysis, deposition, heterogeneous chemistry) and are run at higher temporal resolution.

> *We have added a detailed discussion of our product gas modelling approach explaining the wet deposition within FLEXPART and how the inorganic Bry partitioning takes short-term variations into account. We also added a discussion on the uncertainties of product gas entrainment in modelling and observations. In table 2 we have added observed PG concentrations from ATTREX aircraft measurements in the West Pacific (e.g., Wales et al., 2018).*

The approach to comparisons with the aircraft needs to be improved, both in presentation and approach. Comparisons of the entire observational period or simply a daily mean are insufficient. Comparisons with aircraft data have been done to a higher standard in the literature for many years. Comparisons should at least be made for the same location and time. Why was not a higher model timestamp chosen? The comments that the high concentrations are transient and not captured by models doesn't seem particularly novel, and also likely to be highly sensitive to this.

> *We have improved our model-measurement comparisons by sampling the model output at the measurement time and location for comparisons in Figures 4 and 7. The modelling time step is 30 min and for the campaign periods model output has been saved at this resolution. The new comparison shows slightly different results; however, the overall findings do not change substantially.*

A more general point is that there must be a point at which the limitations from having a single static ocean-surface concentration dataset becomes a limiting factor in what science can be done. Are seasonally resolved predictions really possible without some exploration of the effect of seasonality in the initial concentrations? An opportunity seems to have been missed here to study the impact of perturbing this source of uncertainty.

We agree that the static ocean surface concentrations are currently a limiting factor for assessing the impact of oceanic halocarbons on the stratospheric VSLS entrainment. We have added a discussion of this limiting factor to our manuscript. A recent study from Fiehn et al. (2018) has started to investigate the potential impact of seasonally resolved emissions for the Indian ocean region. While such studies provide valuable insight into potential advantages of using seasonally resolved inventories, it is important to notice that they also have some disadvantages. Fiehn et al. used ocean biogeochemical model simulations to derive the seasonal changes, which did not include production from macro algae, but only phytoplankton sources. Given the limited spatial and temporal coverage of the available measurements, it is currently not possible to derive observation-based seasonally resolved emission inventories.

The language used throughout the manuscript could often be improved. Standard manuscript preparation rules are not followed (e.g. abstract starting with some context for the work). The text requires updating and proofreading for grammar and clarity of language.

We have improved the quality of the manuscript by following standard manuscript preparation rules such as starting the abstract with some context of the work. We have also corrected the manuscript for typos and unclear phrases and sentences.

I cannot recommend publication of this manuscript in ACP without substantial updates as highlighted above and detailed below.

Specific comments

∼Pg. 1 - Please include the word "past" or "historical" in the title and relevant points in the text. This means that the readers will more easily understand how the paper's variability fits with other recent work in the literature, including from the same authors (e.g. Ziska et al 2017).

As suggested, we have added the word 'past' to the title. Furthermore, we have added 'past' or the phrase 'for the time period 1979-2013' to the abstract and text.

∼Pg. 2 Ln 3 - Please start the abstract with context and motivated the work, rather than straight away stating what has been done here.

We have improved the abstract by adding some context and motivation as suggested.

∼Pg. 2 Ln 4 - What is meant here by "available observational datasets"? Do the authors mean all observational datasets have been used, if not which ones and why?

Thanks for pointing this out. We have used all freely available aircraft campaign data that we have been aware of. However, we can see how this phrase is confusing and have changed it to "observational data sets".

∼Pg. 3 Ln 21 - Why is the word "currently" used hear when papers from 2006/2011 are cited. The authors are correct to point out that the heterogeneous chemistry and cycling of bromine has seen notable study recent. Please update this section based on recent literature (e.g. Fernandez et al 2014, Schmidt et al 2016 etc etc)

Thanks for pointing this out. We have updated the references used here based on recent literature.

~Pg. 5 Lns 16 - As before. Please clarify what is meant by “available observational data sets”.

We have changed the phrase to ‘available observational data sets derived during upper TTL aircraft campaigns …’.

~Pg.5 Lns 17 - The use of the terms “high-resolution” to describe 1x1 modelling is odd. This is not enough to really resolve the coastal regions that huge gradients in emissions of the species studied here are seen. Global models are now regularly being run at higher resolutions, such as at horizontal resolution 12x12 km (e.g. Hu et al 2018).

We have changed the phrase to ‘Lagrangian model simulations”. The trajectory simulations are done at a higher resolution than 1°x1° with around 20 million trajectories after the spin up phase. It is true however, that the high-resolution model simulations are limited by the resolution of the meteorological input data given on a 1°x1°.

~Pg. 6 Lns 3-4 - please include the word “static” to ensure the reader is aware that no temporal information is present in the concentrations. This is mentioned later in this section, but should be said at the beginning too.

We have changed the sentence to ‘Static global surface concentration maps of the two compounds were generated from atmospheric and oceanic surface ship-borne in-situ measurements collected within the HalOcAt …’.

~Pg. 6 Lns 12-13 - The gap-filled dataset is therefore 6 years old? Have any measurements been made since that point? Specifically, in locations where effectively no observations are present (e.g. Indian ocean). Would these observations decrease uncertainties within the dataset? What are the uncertainties within the dataset for these regions? Would this mean there is enough data to get a seasonally-resolved dataset?

A recent update of the bottom-up $CHBr_3$ and CH_2Br_2 Ziska et al emission estimates has been provided by Fiehn et al. (2018) after the here presented Lagrangian simulations had been finished. The update shows enhanced emissions in the tropical Indian Ocean and subtropical northern Atlantic by taking new oceanic cruises (Fiehn et al., 2017; Hepach et al., 2016) into account. Globally, the updated emissions are around 10% and 13% larger than existing observation-based estimates for $CHBr_3$ and CH_2Br_2, respectively. We have added a short discussion of the new available data to section 3 pointing out possible consequences for stratospheric entrainment. Given the limited spatial and temporal coverage of the updated observational data set it is not possible to derive seasonally resolved emission inventories.

~Pg. 8 Ln 10-12. Please add a brief discussion of the lifetimes of these species to the introduction. Also state here why the different run periods were used.

We have added information on the atmospheric lifetimes of both gases to the introduction. This information is also given in the section describing the simulation of the chemical decay in FLEXPART. In order to analyse the spatial-temporal variability of stratospheric $CHBr_3$ injections a longer run-time was used. This information has been added to the manuscript.

~Pg. 8 Ln 16. Why is the model run period outside the observational constraint of the emissions (stated as 1989-2011 in the text)? This should be stated in the text.

As we are interested in long-term variations and trends of $CHBr_3$ transport and stratospheric injection, we used a longer model run period than the observational constraint period. We have extended the Ziska et al. (2013) approach by assuming constant $CHBr_3$ concentration fields over the full 1979-2013 time period and calculated the emission fields based on ERA-Interim date over this time period. This information has been added to the manuscript.

~Pg. 9 Lns 6-8 - What about in FLEXPART? Does this reproduce what is seen in TOMCAT? Even with the differences in timestep and online processing?

Yes, the shape of the $CHBr_3$ and CH_2Br_2 profiles (mainly determined by atmospheric lifetimes and transport) are very similar in TOMCAT and FLEXPART simulations, see Figure 1e and 1f in Hossaini et al. (2012) and Figure 12 in our manuscript. Absolut values are slightly different given the different emission scenarios used.

~Pg. 9 Lns 9-10 - What "photochemical" loss? Do the authors mean the prescribed loss terms they have used? What about loss through oxidation (e.g. OH)?

Yes, this refers to the prescribed chemical loss terms. We have added a short statement to the sentence. The chemical loss of $CHBr_3$ is largely driven by photolysis, while CH_2Br_2 loss is driven by oxidation (Hossaini et al., 2010, Figure 4).

~Pg. 9 Lns 11-13 - This approach to Bry cycling and loss is much simpler than what is employed in the state-of-the-art chemical transport models. What sensitivity studies have been performed to see the sensitivity to this? This seems likely to cause large differences in the product gases delivered to the stratosphere.

We also added a general background discussion on the uncertainties of product gas entrainment from modelling and observations. Furthermore, we have added results from a sensitivity study showing how changes of the Bry partitioning impact the product gas entrainment. In addition, we have added a comparison the observational data in order to discuss caveats of our offline approach.

~Pg. 9 Lns 17-23 (I) - These authors have not convinced the reader that the partial use of offline partitioning is appropriate. The lifetimes of some of these inorganic species are of the order of minutes and partitioning dramatically changes from day to night. Why too are partitioning calculations being used from 10-15 years ago, have the values predicted by the field not changed? What are the differences between the simulations of inorganic bromine in these simulations and those (even in the same model) in more recent work (e.g. in CAM-Chem, GEOS-Chem, and TOMCAT)? Either strong evidence that the answer is insensitive to this simplification needs to be added, or the discussion of product gases should be removed.

The 3-dimensional Br_y field from p-TOMCAT and its partitioning into HOBr, HBr, Br, BrO, $BrONO_2$, and Br_2 are given with a temporal resolution of 30 min. As the partitioning of the Br_y field varies strongly with location and time, we apply it in a first step to every air parcel according to its location each time before the wet deposition is initiated. In a second step, wet deposition is calculated individually for each inorganic bromine species based on its solubility specified by the effective Henry's law coefficient. Once wet deposition is initiated the Br_y fraction determined to be washed out is removed completely. This information has been added together with further details to the manuscript.

The simulations are not insensitive to the specific partitioning used in the study in the same way that online chemistry simulations can result in different wash-out rates. Our citation of the Yang et al. (2005) is misleading. We use a chemical partitioning derived from p-TOMCAT runs from 2012 which are based on the known homogeneous bromine chemistry and include heterogeneous reactions on aerosols (reactivating bromine radicals from the reservoir species) and we have added the relevant Yang et al. (2010) citation. We apply the partitioning every 30 min in order to take into account the high variability and day-night differences. Uncertainties in the modelled wet deposition arise from the parameterization of solubility via the effective Henry's law coefficient and from uncertainties in the Br_y partitioning caused by errors in the aerosols loading and in the mechanism used for heterogeneous reactions. Clouds and aerosols within p-TOMCAT are not matched with those in FLEXPART which might lead to an additional error source. A short discussion has been added to the manuscript.

~Pg. 9 Lns 17-23 (II) - Please give a statement here about how the model has been evaluated against the observations for inorganic bromine (like a similar statement for the VSLS on Pg. 8 Ln 30-32). Multiple recent studies have reported these observations (e.g. Koenig et al 2017)

We have added a table to the manuscript which compares VSLS source and product gases derived from measurements of the ATTREX and CONTRAST campaigns to estimates derived from the FLEXPART model simulations. The comparison for the West Pacific region shows good agreement for both, SG and PG entrainment. Note that the Br_y derived from measurements is based on BrO measurements and chemical box model simulations.

~Pg. 9 Lns 17-23 (III) - More explanation needs to be given of how the mixture of the online and offline process are considered here? There is a prescribed lifetime, does this just include photolysis or oxidation too? But it doesn't include wet deposition? Or are depositional losses being double-counted here?

The prescribed lifetime of $CHBr_3$ and CH_2Br_2 includes photolysis and oxidation but no depositional loss. The washout is only applied to the inorganic Br_y. A more detailed explanation has been added to the manuscript.

~Pg. 9 Lns 25-36 - Why not diagnosis the tropopause from the ECMWF met fields? This seems like an unnecessary simplification.

We used the cold point tropopause diagnosed from the ECMWF fields, which was calculated within the FLEXPART routine. We have added this information to the sentence.

~Pg. 10 Lns 3-5 The authors are right to point out this large uncertainty on their prediction of product bromine. However, a full discussion of this is essentially missing from the manuscript and must be added.

We have added a more detailed discussion of product gas entrainment based on our sensitivity studies and based on the comparison to the measurement derived Br_y estimates. The fact that source and product gas entrainment of $CHBr_3$ and CH_2Br_2 agree relatively well with currently available measurement-based estimates and with other model estimates indicates that the simplifications applied in our modelling approach (lifetimes of both gases and Bry partitioning) are consistent with the current state of knowledge.

~Pg. 17 Lns 1-11 What other model-observation comparisons have been done in the literature. Have these failed or succeed in capturing the observations? Please add this information to give context for the work.

According to the best of our knowledge there is no model evaluation or comparison that has used the ACCENT data.

~Figure 2 - Why do units of concentration presented in Fig 2d in have a time dimension?

Thanks for pointing this out. We have corrected the units in Fig. 2d.

~Figure 3 - Please explain the noisy pattern seen in the model values underlayed.

The spotty features in the model simulations are a result of the high oceanic sources directly underneath interacting with localized convective transport. The latter brings localized air masses with very high bromoform mixing ratios from the boundary layer into the 15-17 km layer. We have added this explanation to the manuscript.

~Figure 4 - This figure does not clearly convey information. Why are whole campaigns solely represented as stars? Why not show some level of variability seen by both the modelling and observations? It seems like a simple box and whisker comparison would convey more information here. Why two are the multiple horizontal lines, is this a split y axis? If so please show this with an axis break symbol.

We have improved the figure by including the measurement variability and by adding an axis break symbol. Furthermore, we have improved our model-measurement comparisons by sampling the model output at the measurement time and location.

~Figure 7 - Please show uncertainties or ranges for the observations from ATTREX on the leftmost and rightmost subplots. Also, the paradigm for plotting in atmospheric science is usually that observations are shown in black and model. Please update this plot and anywhere else in the manuscript to avoid confusing readers.

We have improved the figure by including the ATTREX measurement variability and by showing the observations in black. Furthermore, we have improved our model-measurement comparisons by sampling the model output at the measurement time and location.

~Figure 8. (e) Please explain discontinuity seen at latitudes of -5 degrees N and 5 degrees N in the Indian ocean. These do not seem physically plausible.

These discontinuities result from the emission inventory which shows similar patterns in the same region. We have added a figure of the seasonal emissions to the supplement.

~Pg. 37 Ln 1 - Are only the bromoform emissions archived? What about the dibromomethane, where is this data archived?

Thanks for pointing this out. The dibromomethane data is also archived and we have updated to manuscript accordingly.

Technical comments

~Pg. 2 Ln 22 - expand “Br/dec” to include “decade”.

We have changed the manuscript accordingly.

~Pg. 5 Ln 5 - Typo - “meteorological”

We have corrected the typo.

~Pg. 5 - Ln 23-24 - Sentence does not scan well. Please rephrase. “The question if such hotspots are mainly driven by oceanic or by atmospheric processes will be answered based on the Lagrangian simulations.”

We have changed the sentence to “We will investigate if such hotspots are mainly driven by oceanic or by atmospheric processes by analysing emission patterns and transport pathways derived from the Lagrangian simulations.”

~Pg. 8 Lns 4-7 - Please update the sentence as below for readability. The sentence structure here and elsewhere could be more concise. From “with the Lagrangian particle dispersion model FLEXPART (Stohl et al., 2005) 7 Version 9.2 beta.”To: From “with the FLEXPART Lagrangian particle dispersion model (Version 9.2 beta; Stohl et al., 2005).”

We have changed the sentence as suggested.

~Pg. 11 Lns 25-27 - “In the following,” is verbose and should be removed.

We have removed the phrase.

~Pg. 35 Ln 18 - Add spaces between numbers and units here and elsewhere in the manuscript. Please refer to NIST guidance on this (https://physics.nist.gov/cuu/Units/checklist.html).

We have added the missing spaces between numbers and units.

~Pg. 11 Ln 25 - Both analyze and analyse are used in multiple places in the text. Please choose to use either the American or British version.

We have changed the manuscript and use the British version.

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