

Review of:

Impact of a moderate volcanic eruption on chemistry in the lower stratosphere: balloon-borne observations and model calculations

by G. Berthet et al.

Anonymous Reviewer 2

1 General Comments

The manuscript presents a measurement model intercomparison study of the effects of the aerosol plume of the Sarychev volcano eruption in June 2009 on the high latitude lower stratospheric nitrogen, halogen and HOx chemistry and the relevance to the lower stratosphere ozone budget. The study employs a broad spectrum of balloon-borne measurements of aerosols and trace-gases as well as several sets of satellite aerosol data. The model employed is the REPROBUS 3D CTM that is well-known from several studies mainly focussing on process understanding. Here the focus is on the effects of the plume of a moderate volcanic eruption (as opposed to several studies on major volcanic plumes as Pinatubo) which is worthwhile since a series of these moderate eruptions is supposed to be one cause of the long-term increase in the global stratospheric aerosol density. Several interesting aspects of the chemistry are presented and special features of the combination of this moderate eruption plume with high-latitude lower stratospheric chemistry are identified. Therefore a general interest for such a study is clearly given and ACP represents a well-suited platform for the publication.

However, the reasoning for exploring the effect of a moderate eruption on chemical composition at Arctic latitudes in summer must be better rationalized in the introduction. Why is this important? Currently only the relevant chemical, aerosol and some dynamical processes are explained.

From the satellite data presented in Haywood (2010) and Jegou (2013) and also Fig.5 it seems obvious that the plume is not homogeneously mixed over the Arctic region by August/September 2009. Therefore the expected horizontal and vertical structure has to be discussed in some more detail. The STAC balloon data from different flights needs to be introduced in Fig. 1 (e.g. grey underlayered traces?), not just

ranges of the observations so the reader gets a better impression of the vertical structure of the plume and its variability. Horiz. and vertical variability should be discussed at least. Currently no filtering for high/low or even background aerosol regions is done for the interpretation of the data which may well be warranted but must be better supported.

In order to explore the sensitivities of the model study to different parameters such as differences in aerosol surface area or dynamical effects a number of differently constrained simulations have been carried out and are intercompared in the figures and tables presented. Therefore partly the figures and tables and consequently the discussion gets quite busy and confusing. The results for the runs termed sat-sim and bal-sim generally don't differ by more than 10%, mostly much less. Therefore this just needs to be shown in one plot (Fig. 6) but then can be neglected just stating that differences for other species are also minor. The same is true for the 1D simulations which are meant to check on dynamical influences. Once the results of these model experiments have been stated the following discussions can be simplified a lot by leaving all the other simulations out. Especially Table 2 should be considerably simplified, I'm not aware that all the various differences presented there are even discussed in the text.

The balloon-borne measurements acquired during the Strapolete experiment represents a data set that nicely covers an interesting episode of aerosol enhancement in this atmospheric domain and therefore publication is of its own value. Possibly a link or links to the appropriate data base(s) should be also given in order to enable use of the data for other studies.

In general I think the manuscript should be published, however, after considering the general and detailed comments and streamlining the presentation and discussion.

2 Detailed and minor Comments

p4:l16 Is the wording “We focus here on ...” meant to discriminate against the aerosol measurements or does it just refer to gas-phase data?

p4:l16ff Why are there only ascent data used for the SPIRALE measurements? It would be interesting to see also descent data to get a feeling on variability and possible contamination issues on ascent since the cell extends below the payload.

p5:l3ff This paragraph is somewhat chaotic and hard to understand and should be polished. Before switching to the BrO profiles a new paragraph might be started.

p5:l21ff The fact that REPROBUS is used without any detailed sulfur chemistry should be clearly stated right away then referring in which different ways the aerosol plume is prescribed.

p5:l53 The Haywood (2010) reference is missing.

p6:l33 The analytical expression for the derived correlation in Fig.3 should be given.

p6:l37 Better leave out the phrase *in the model*.

p9:Sect.3.4 can be considerably shortened since it doesn't add new results (see Fig.9).

p9:l53 Why is the nomenclature changed here to Balloon-aero-sim instead of Bal-sim etc.?

p10:Sect.3.6 The stated improvements of the 1D simulations above 20km are not at all obvious to me and are certainly not significant improvements that can be employed for the conclusion drawn in this section.

p12:l10ff With the introduction of Fig. 11 the dramatic difference of the two BrO profiles should be explained (sza?). Also what is the tropopause height for the DOAS measurement? More than 1 ppt of BrO below the tropopause seems quite suspicious to me.

p12:l35 The meaning of the percentage changes for switching off the BrONO₂ hydrolysis must be more clearly explained. For the example given it should be 16% of the daytime BrO production not 18%.

p12:l39 This result implies ...

p12:l52 ... the active chlorine family species ...

p13:l30ff I propose to use absolute values for the ozone loss discussion (see the comment on Fig.12).

p13:l39 ... into the lower stratosphere.

p15:l33 When switching to absolute values for the accumulated ozone losses the discussion why “largest” losses occur just above the tropopause will become obsolete, I guess.

p16:l35 It might be interesting ...

p18:l19 Does the *uncertainty* represent accuracy? Since several balloon-borne measurements are used along with each other and are compared to other aerosol SAD data it is not sufficient to just state the precision of the measurements.

p18:l41 I guess *overall uncertainty* represents accuracy? Rename or otherwise state the accuracy.

p19:l17 The Voigt et al. reference is missing. Also the Pfeilsticker et al. reference (120). Please check over completely!

p21:l3 ... strong functions ...

p22 All citations should be thoroughly rechecked since several citations have been missing from the references list.

Table 2 Several of the tabulated differences are not used at all in the text. The table can be reduced considerably or even removed completely.

Table 3 In the caption it should better read: Numbers are taken from the Sat-sim simulation. Also ... BrONO₂ hydrolysis (Reaction 4) to changes ... would help.

Fig.1 I propose to show the traces of the individual measurements on the plot to give the reader an impression on the variability. Also the altitudes shown should be extended somewhat to 25km and possibly below 10km to give a better impression on the vertical extend of the plume.

Fig.2 For my taste in the right panel SPIRALE HNO₃ should be included.

Fig.5 The grey shaded (error bars?) on the NO₂ profile should be explained in the caption.

Fig.9 Due to the high partitioning into HNO₃ this species is not sensitive for the aerosol effect. This plot therefore can be left out.

Fig.11 The solar zenith angles of the profiles have to be given in the caption due to the diurnal variation of BrO.

Fig.12 Showing the percentage changes in ozone is somewhat misleading. In order to point out the altitude regime of highest impact in terms of ozone loss an absolute scale should be used like loss rates of ppb/day. a percentage change of 5% doesn't have any major effect if ozone levels are negligible at the TP.