- We thank Mr. Fromm for his comments (in red). 1
- The title is specific and restrictive to tropospheric water vapor. This, and the fact that their Part I 2
- paper is similarly entitled, implies intent to limit the scope thusly. However the body of the paper 3
- includes stratospheric as well as tropospheric water vapor observations. E.g. Page 25881, L14-4
- 25; P25885, L4-5. If the authors' intent is to reflect the title, the entire stratospheric part of the 5
- paper is out of scope. Otherwise the titles and motivation of both papers need to change. 6
- 7 Our intent is not to only reflect the title with every sentence of the manuscript. The title cannot
- contain all of the intended material in a paper. Based on comments by the other reviewers, the 8
- Nabro section has been deleted, meaning that no part of the paper is out of scope. The discussion 9
- phase for the companion paper is closed so Mr. Fromm is out of place and out of time to 10
- 11 comment on that paper.
- Regarding VEI No citation is given for the VEI construct. 12
- We now cite Newhall and Self (1982). 13
- VEI is not discussed in the cited Smithsonian report for Puyuhue Cordon Caulle. 14
- VEI is quoted under the "Eruptive History" tab of the website provided. This information was 15 present at the time of submission of this manuscript. 16
- 17 The manuscript is not changed based on this comment.
- Moreover, VEI is qualitatively proportional to injection height, with VEI of 5 or more being 18 strictly stratospheric. Of what relevance is a 5+ VEI to upper tropospheric water vapor? 19
- VEI is qualitatively proportional to injection height but this statement is not complete. VEI is 20
- based on the volume of ejecta and the column height. A larger VEI implies a larger volume of 21
- water emitted into the atmosphere as a whole as well as greater capability to entrain ambient 22
- water vapour in the lower atmosphere. When a volcano erupts to a height of 15 km, for example, 23
- not every emitted or entrained molecule reaches 15 km. This is especially true for water vapour 24
- which condenses as it rises through the troposphere. 25
- The manuscript is not changed based on this comment. 26
- The abstract gives information that is found nowhere in the paper and which is incorrect: that the 27
- Cordon Caulle eruption was "the most explosive eruption in the past 24 years." Clearly several 28 29
- volcanic eruptions since 1991 have been more explosive, including Pinatubo.
- 24 years was obtained by subtracting 2015-1991 with the eruption of Cerro Hudson (August 30
- 1991, VEI=5) defining the start of this 24 year period. Our manuscript was accepted into ACPD 31
- on 6 September 2015 and there have been no major eruptions since then so the statement is still 32
- accurate. Pinatubo erupted in June 1991. There were later significant eruptions (with heights of 33
- 10 km) until September before the 1991 Pinatubo eruption eventually stopped. We note that the 34
- 2015 Calbuco eruption has not been assigned a VEI to date. In any case, the period from 35
- September 1991 to April 2015 rounds up to 24 years. 36

- 1 No change to the manuscript is necessary based on this comment.
- 2 The authors inexplicably ignore the high-latitude Grimsvotn (Iceland) eruption of May 2011.
- 3 The Grimsvotn material was in the UTLS at high latitudes even before Nabro woke up. It would
- 4 seem that any discussion of volcanoes and UT water vapor at high northern latitudes in 2011 has
- 5 to involve Grimsvotn, which had both a head start and preferable latitude w.r.t. Nabro.
- 6 The opening sentence of Sect 3.2 clearly states that July 2011 had little indication of enhanced
- 7 water vapour in the northern high latitude upper troposphere. This eliminates the local eruption
- 8 of Grímsvötn as the major contributor in September 2011. At 5.5 km (the lowest available
- 9 altitude level for ACE), there are only three years in which MAESTRO has a significant sample
- size in May and no such years for ACE-FTS due to perpetual, optically thick clouds. So not
- 11 much can be said about Grímsvötn at 5.5 km and the three available years from MAESTRO are
- all within $\pm 3.5\%$ of the average of the three. Above 5.5 km, there is no suggestion of a
- 13 significant positive anomaly (>6% and >1 standard deviation large than climatology) in northern
- high-latitude MAESTRO water vapour data that persists from May 2011 to July 2011 at any
 altitude (6.5-19.5 km).

16 No change is made to the manuscript based on this comment since the normality of July 2011 17 had already been discussed in Sect. 3.2.

18 Regarding "recent eruptions such as Kasatochi" (in the paper's wrap-up section) the authors

- 19 claim that these other eruptions had little impact on stratospheric water vapor. Several issues
- with respect to this: 1. the authors presented no analysis of these other eruptions, 2. they give no
- citation, and 3. the stratosphere is of questionable relevance to the theme of upper tropospheric
- 22 water vapor.

23 1. We presented only the eruptions which most obviously perturbed high-latitude UTWV.

- 24 Puyehue was the only eruption in the southern hemisphere that was outstanding. We looked for
- 25 monthly median relative anomalies that ranked first in terms of magnitude at a particular height
- as compared to other positive anomalies from all months of both MAESTRO and ACE-FTS high
- 27 latitude data records. This led to the selection of Puyehue (July 2011, 8.5 km), Nabro (Sep 2011,
- 28 12.5-13.5 km), and Eyjafjallajökull (May 2010, 9.5 km). No other eruption met this criterion.
- 29 The highest ranking negative anomalies did not appear to coincide with volcanic activity. Using
- 30 this criterion essentially means that any other volcano did not enhance water vapour by \sim 50%
- over a month or in one case (6.5 km, July 2008, possibly related to Okmok), that only one
- instrument (MAESTRO) was able to see this low altitude frequently enough, so we chose to skipit.
- 34 2. This sentence has been deleted.
- 35 3. Mr. Fromm is correct that this statement is of questionable relevance to the main theme of thepaper.
- 37

1 <u>Response to reviewer 1</u>

2

3 We thank the reviewer for their valuable comments (in red).

- 4 This paper makes a case that volcanic eruptions may significantly enhance the amount of water
- vapour in the lower stratosphere. (...) By correlating aerosol extinction measurements with water
 vapour measurements they conclude that the water source is from the volcano even though the
- 7 measurements themselves occurred a few months after the eruption.
- 8 Puyehue erupted several times with the plume reaching 9 km above the crater on June 13, 2011
- 9 (http://volcano.si.edu/showreport.cfm?doi=GVP.WVAR20110608-357150) and circumnavigated
- the globe twice before the end of June 2011 (see cited Vernier et al., 2013). The ACE
- 11 measurements of this plume in Fig. 5 start only 18 days after eruption. Nabro finished its major
- eruptive phase on 14 June 2011 with plume height in excess of 10 km
- 13 (http://volcano.si.edu/volcano.cfm?vn=221101). ACE measurements following Nabro begin on
- 14 July 2nd, 2011 (Fig. 10), only 18 days later, which is actually too early in the sense that the plume
- had not reached all longitudes by this point (Bourassa et al., 2012). As written in the paper,
- 16 Eyjafjallajokull began its major eruptive phase on April 14th, 2010, but there was a second major
- eruption on May 5^{th} that reached ~10.0 km (refer to the cited Gudmundsson et al., 2012) and the
- 18 first ACE high latitude measurement occurs 11 days after this second eruption and sampled the
- volcanic plume. So in all cases, local ACE measurements first become available roughly two
- weeks after a major eruption. Despite this good fortune, ACE does not have the spatiotemporalcoverage of limb emission sensors.
- 22 We have inserted the following sentence near the start of section 3.3:
- This was followed by a second eruption on 05 May 2010 that also reached ~10.0 km
 (Gudmundsson et al., 2012).
- For the most part the analysis is straightforward and reasonable but there is an issue that also need to be acknowledged. Referring to the Schwartz et al. GRL, 40 2316-2321,
- doi:10.1002/grl.50421,2013 paper, it turns out that 2010 and 2011 were years where convective
- 28 injection of water vapor was quite active and intense, producing events as high as 18 ppmv
- 29 against a background value of 5 ppmv.
- 30 Somehow we missed this paper in our literature search (and hopefully not other relevant ones)
- and apologize to the reviewer if they were an author of this excellent and highly relevant paper.
- 32 Schwartz et al. (2013) show that in summer 2010 and 2011, MLS appears to observe water
- vapour mixing ratios of \geq 7 ppm more frequently than other years. This is the only metric used by
- 34 Schwartz et al to illustrate interannual convective differences. They are omitting the small
- possibility that Nabro may have contributed to the more frequent extreme water vapour VMRs in
- 36 2011 (but not 2010 obviously).
- 37 Our analysis of northern mid-latitude water vapour with MAESTRO (which lacks the spatial
- 38 coverage of MLS) indicates that at altitudes corresponding to 100 and 82.5 hPa, water vapour
- 39 VMRs are not enhanced when averaging over two summer months (namely July and September

1 2011, Fig. 10) and we used the same span of years as Schwartz et al. (2004-2012). In fact, at all

- 2 altitudes in the range 8.5-19.5 km except for the 1 km thick layer centered at 13.5 km,
- 3 MAESTRO measured normal or below normal water vapour VMR in these two months. It is
- 4 quite possible that 2011, for example, can have a higher frequency of extreme water vapour
- 5 VMRs without having a higher central tendency (e.g. median) than other years because the
- 6 extreme events are too rare to significantly affect the zonal monthly median or even the mean. It
- 7 is also possible that MAESTRO has a dry bias because we do not observe when optically thick
- 8 clouds are present. MAESTRO can see through thin cirrus however and so the dry bias is
- 9 probably largest below 12 km and vanishing up toward 17 km. MLS may also have such a dry
- 10 bias for clouds with large drops/crystals. In any case, what we present in our manuscript is
- 11 mostly a statistical analysis. We cannot prove without doubt whether or not the water vapour
- 12 enhancement in the northern high-latitude lower stratosphere could have occurred without
- 13 Nabro.

As can be seen in the response to Reviewer 3, we credit Reviewer 1 with providing an alternate process (i.e. summertime deep convection) responsible for the enhancements in water vapour in the northern extratropical tropopause region and we have removed the Nabro section.

- 17 Even though the air may be aerosol enriched, by virtue of the two month or so time lag, it is
- possible if not probable, that the moisture in these air parcels could be enriched by convectiveevents. I think this possibility should be acknowledged.
- 20 We acknowledge that during the period of study for Nabro (specifically July-Sept 2011), deep
- 21 convection was likely the main mechanism for the ACE-observed enhancements in zonal median
- 22 water vapour in the vicinity of the northern extratropical tropopause. Convective events in
- 23 Uruguay (Schwartz et al., 2013) during their winter are expected to be rare and this expectation
- is confirmed by Schwartz et al. Puyehue was the most likely cause of the July-August 2011
- 25 enhancements, given the explosivity of its eruption. We do not believe that the reviewer's
- 26 comment pertains to Eyjfjallajökull since that eruption was in the spring and outside of the
- summer period in which the anomaly in deep convection was observed.
- The Nabro section has been deleted. No change was made to the sections on the other twoeruptions.
- 30 A water vapor enhancement signature should be evident shortly after an eruption, even if it is
- 31 injected as ice on particulates because the stratosphere is of very low humidity and sublimation
- 32 should occur rapidly. I appreciate, that occultation type instruments do not sample well enough
- 33 to capture a plume early in the eruption cycle. Even instruments like MLS or MIPAS often miss
- 34 plumes in their early stages, but it would be worth looking at their data to see if enhancements
- are seen as they should produce bigger signatures and contrasts against background amounts.
- 36 The main focus of this paper and the companion paper is to understand water vapour variability
- 37 on timescales of one month or longer for the high latitude upper troposphere as a function of
- altitude (5 to \sim 10 km). This focus is clearer in the revised manuscript with the discussion of
- 39 stratospheric injection of water vapour now removed and the Nabro section deleted. MLS
- 40 measurements do not go low enough to cover the entire 5-10 km altitude range. As discussed in

response to the next comment, we are not looking for bigger enhancements in individual 1 observations and we are not contrasting against the local background. We are contrasting in time, 2 comparing e.g. the month of September 2011 at northern high latitudes to other Septembers in 3 this same region and looking for "big signatures" on monthly timescales. However, we share the 4 reviewer's interest in volcanogenic perturbations to stratospheric water vapour, so we looked at 5 MLS water vapour on the day after the Nabro eruption, namely June 14, 2011. We were guided 6 to MLS observations of the Nabro plume by Fig. 1 of the work of Fromm et al. (2013) entitled 7 'Comment on "Large volcanic aerosol load in the stratosphere linked to Asian monsoon 8 9 transport"' (Science 339: 647, DOI: 10.1126/science.1228605). Figure 1 below shows MLS v4.2 10 SO₂ at 100 and 68 hPa as a function of latitude for an orbit intercepting the Nabro plume. The highest SO₂ VMR at 100 hPa occurs at a latitude of 26.7°N with the second highest value being 11 12 in the adjacent limb scan at 25.2°N. At these same adjacent latitudes, MLS v4.2 water vapour from the same orbit is clearly enhanced at both 100 hPa and 121 hPa, but not at 82 hPa (Fig. 2). 13 This indicates that a 2 ppm enhancement in water vapour exists at 100 hPa at a latitude of 14 26.7°N, but not at altitudes above that. Meanwhile, SO₂ appears to be enhanced at 68 hPa as 15 well, particularly at 25.2°N. The tropopause pressure according to the MLS v4.2 temperature 16 product indicates the tropopause pressure was 100.3 and 99.9 hPa for the two adjacent latitudes 17 of note. This evidence from MLS suggests that a significant amount water vapour was not 18 directly injected into the stratosphere by the Nabro eruption. However, the information presented 19 20 here does not rule out that water from Nabro could have been directly injected in other phases 21 (e.g. ice). We did not try using MIPAS data given that MLS orbit of observations were very clear

(e.g. te), we did not try using Wit AS data given that MLS orbit of observations were very clear about the altitude range and magnitude of the water vapour and SO₂ VMR enhancements on June

23 14, 2011.

There is no change to the manuscript because the Nabro section has been deleted. The other two

25 studied volcanoes do not appear to be addressed by this comment.



 $\ \ \, \text{Figure 1} - \text{MLS SO}_2 \text{ VMR at two pressures (hPa) at low latitudes for an orbit intercepting the}$

3 Nabro plume on June 14, 2011.



2 Figure 2. MLS water vapour VMR at three pressures (hPa) at low latitudes for an orbit intercepting the Nabro plume on June 14, 2011. 3

A case in point being in the Discussion (page 25885, line 10) that Kasatochi produced little 4 impact on stratospheric water. MLS did observe enhancements in H₂O from this eruption (see 5 Schwartz, 2013 for reference); hence, the other volcanoes should produce even bigger signatures 6

7 near eruption if they are able to influence the stratospheric water vapour budget as claimed.

8 We did not claim that Puyehue and Eyjfjallajökull influenced the stratospheric water vapour 9 budget. Nabro was a tropical volcano and the tropopause is very high in the vicinity of the 10 eruption leading to larger background volume mixing ratios at ~13.5 km, the altitude of the ACE-observed water vapour enhancement at mid-latitudes in July and September 2011 (Fig. 10). 11 The reviewer's comment implies that in order to influence the stratospheric budget, volcanoes 12 must produce big water vapour 'signatures' near the eruption. Our original paper hinted at a 13 14 second mechanism that again does not require direct stratospheric injection that ironically is the main focus of Schwartz et al. (2013): the monsoon (specifically the Asian one). In the revised 15 manuscript, after a more probing and latitudinally resolved analysis (including examination of 16 individual profiles), we no longer claim that Nabro influenced the stratospheric water budget. 17 18

A last point, even the southern hemisphere is also affected by mid-latitude convection events like 19 20 those in the north (usually occurring over Uruguay) but they are not as frequent or intense.

- Mid-latitude convective events are expected to be less frequent and less intense in winter (e.g.
- July 2011), when Puyehue water emissions were detected in the upper troposphere. For Puyehue,
- we find water vapour enhancements at 6.5 to 9.5 km. Thus, the convective events at 100 hPa
- (~16 km) illustrated in Schwartz et al. are not very relevant. The spatial pattern of water vapour
- at 6.5 km could be much different.
- At p25884L25, we have rewritten the original sentence as follows:
- Volcanic UTWV enhancements in the extratropics during the cold season are more readily
- detected in monthly zonal median data because of the low background VMR of water vapour in this region and season, owing to the lack of deep convection.
- Minor correction, page 25875 line 14 should 2002 be 1992?
- Thanks to the reviewer for noticing this strange mistake. It has been corrected.
- Page 25879 line 7-8, you talk about Austral summer and also July / August. This is Austral
- Winter.
- We thank the reviewer for catching this. This has been corrected (twice).

Response to reviewer 3 1 2 We thank the reviewer for looking closely at all aspects of this paper. In a nutshell, we have 3 4 reorganized the paper to make the purpose of the study clearer. We have also completely removed the section on Nabro, given that the enhancement appeared to be tropospheric even at 5 mid-latitudes and thus was not large enough in magnitude to persist following transport to 6 northern high latitudes, given the short residence time of UTWV, particularly for mid-latitude 7 summer. 8 9 10 I think the authors address an interesting question but the presentation is confusing and the physical argumentation unclear, such that I could not really follow the line of thoughts. 11 12 13 We agree that the presentation could be more clear. We try to improve the connection between successive thoughts. We have also removed the Nabro section (see below). 14 15 We now write: 16 17 18 (p25879L11) To connect the clearly enhanced UTWV at southern high latitudes to the eruption of Puyehue-Cordón Caulle (Puyehue hereafter), UTWV profiles in the 40-60°S band, which 19 contain the latitude of this volcano, were contrasted between July 2011 and July 2012 (a normal 20 July). Figure 5 shows a statistically significant increase in UTWV in the 40-60°S latitude band as 21 well for July 2011 relative to July 2012, and no significant increase above 10 km. 22 23 and add: 24 25 (p25879L16) "...in July 2011..." 26 27 28 and change: 29 (p25880L1) "is advected to" -> "resides in" 30 31 I recommend a complete rewriting of the paper after the authors have carefully reconsidered how 32 33 they think that volcanic emission can impact upper tropospheric humidity in remote areas on time scales of several weeks. 34 35 For Eyjafjallajökull, while the water vapour enhancement in July 2010 at 11.5 km is largest in 36 37 terms of rank for that altitude and calendar month, the enhancement is not significant for MAESTRO relative to the standard error of the July 2010 monthly mean at 11.5 km. Thus, for 38 this eruption, the period of significant enhancement is only May 2010 (during which the eruptive 39 phase was ongoing and within the northern high-latitude band) and thus the timescale is not 40 questionable given the lifetime of UTWV of ~21 days (Ehhalt, 1973), discussed below. 41 42 The last two sentences of Section 3.3 now become: 43 ACE does not sample northern high latitudes in June. In July 2010, enhanced UTWV is observed by both instruments only at the local tropopause (11.5 km), but for MAESTRO, this 44

45 enhancement is not statistically significant.

For Puyehue, in our reply to the comment on Trenberth (1998) below, we show that the UTWV 1

- enhancement observed in July-August 2011 is consistent with the residence time of water vapour 2
- at the tropopause (which was located at 9.5 km averaging over July 2011 for example), and we 3
- now write at p25880L12: 4

13

14

15

5 "whereas in September 2011, the UTWV enhancement is statistically insignificant."

For Nabro, we now believe the conclusion that the enhancement at northern high latitudes was 6

- mostly due to this eruption is incorrect. In both July and September 2011 at northern mid-7
- latitudes, there is a positive anomaly in water vapour of 2 ppmv at 13.5 km. This anomaly is 8
- consistent both in terms of the absolute magnitude and altitude and no other altitude shows a 9 10 significant enhancement in either month.
- The July 2011 mid-latitude enhancement of 2.4 ± 2.2 ppmv appears to be tropospheric after 11
- separating profiles from this month and latitude bin into two groups: 12
 - A) with a tropopause ≤ 13.5 km, and
 - B) with a tropopause >13.5 km

16 Only group B showed a clear RH enhancement at 13.5 km and it was vertically narrow feature

17 (spanning 12.0-14.0 km). Since it appears to be tropospheric, even if it was entirely due to

Nabro, it would be likely reduced to ~0.2 ppmv by mid-September assuming a tropopause 18

residence time of 3 weeks (which may be too long since the tropopause was >13.5 km in 26% of 19 the July 2011 mid-latitude MAESTRO profiles). If this mid-latitude enhancement were to cross

20 the tropopause as it was transported poleward, it would suffer the least depletion if it were 21

- transported to the lowermost stratosphere as early as possible. 22

A stronger argument against Nabro being the main source of humidity at northern high latitudes 23

in September 2011 is that the enhancement at 10.5 km at northern high latitudes is significant for 24 25

both instruments and the absolute magnitude is 5.7 and 10.8 ppmv for ACE-FTS and

MAESTRO, respectively. These numbers greatly exceed the mid-latitude enhancement of ~2 26

- ppm at 13.5 km. This supports the idea that the high-latitude enhancement at 10.5 km was not 27
- primarily due to Nabro since those enhancements appear to be too large to be related to a 2 ppm 28 29 mid-latitude enhancement and a different mechanism (e.g. deep convection) is likely the main 30 one.
- Figure 13 of the original manuscript shows that in September 2011 at northern high latitudes, 31
- MAESTRO sees a statistically insignificant enhancement of 2.7 ppmv at 12.5 km considering 1 32
- 33 km vertical bins and that ACE-FTS shows an enhancement of 1.4 ± 0.9 ppmv at this altitude. We
- removed the single September 2011 high-latitude profile which had a tropopause above 12 km 34
- (Figure 12), which resulted in a reduction of the ACE-FTS monthly median at 12.5 km by 0.01 35
- ppmv. The mid-latitude enhancement in July 2011 does not appear to be sufficient to explain the 36
- 37 September 2011 enhancement at 12.5 km at high latitudes, accounting for a residence time
- appropriate to the tropopause (3 weeks). 38
- Furthermore, examining MAESTRO water vapour profiles from September 2011 in the 30-60°N 39
- band, the enhancement at 13.5 km appeared to be coming from profiles in the 50-60°N latitude 40
- range where there was a sharp gradient in both RH and water vapour VMR at 13.5 km while the 41
- lower latitudes (30-50°N) showed no sign of a vertically confined enhancement at 13.5 km. The 42
- 43 enhancement at 13.5 km in the 50-60°N data appeared to be tropospheric in origin and most

- 1 apparent in profiles with the highest tropopauses (e.g. 13.5 km, compared to the normal value of
- 2 11.5 km). The enhanced monthly zonal median in September 2011 at northern mid-latitudes is
- likely due to deep convection, which pushes up the local tropopause, and results in high water
 vapour VMR and RH near 100% through the entire upper troposphere, as shown in a sample
- 5 profile below (Fig. 1).





Figure 1. RH profile for sr43466 on Sept. 8th, 2011 at 50.5°N derived using MAESTRO water
 vapour.

We believe, as suggested by reviewer 1, that unusually deep convection in the summer of 2011 is
more likely to explain the positive anomaly in the high-latitude tropopause region observed in

more likely to explain the positive anomaly in the high-latitude tropopause region observed in
 September 2011. The northern mid-latitude UTWV enhancements, particularly in September

- 14 September 2011. The normer indefinition of www emancements, particularly in September 15 2011, are also more likely to be due to unusually deep convection (Schwartz et al., 2013) than
- 15 2011, are also more likely to be due to unusually deep convection (Schwartz et al., 2013) than 16 from a low latitude volcano months earlier. Also, MLS observations of the low-latitude Nabro
- 17 plume one day after eruption indicate that enhanced water vapour reached the tropopause but
- essentially did not appear to have gone any higher (see response to reviewer 1), indicating that if
- 19 the volcanic enhancement reached the stratosphere, it would have been during subsequent
- 20 poleward transport, meaning the water vapour enhancement resided initially in the low latitude
- 21 upper troposphere where precipitation could deplete it more quickly.
- 22

2 stratosphere. 3 4 1) General: I found nowhere a good explanation of why this study focuses on water vapour in high latitudes. 5 6 The motivation for studying high latitude UTWV is provided at the end of Sect. 1: 7 8 9 Currently, trends in UTWV are not known for high latitudes (Hartmann et al., 2013). However, the main focus of this work is on improving our understanding of UTWV variability at high 10 latitudes and the role of volcanic emissions relative to other dynamical and thermodynamic 11 processes in this region (see companion paper: Sioris et al., 2015). 12 13 Instead of the second sentence of this excerpt, we now write: 14 15 The first step toward accurate trends is to improve our understanding of UTWV variability at 16 high latitudes. The variability of upper tropospheric water vapour (UTWV) at high latitudes is 17 dominated by dynamics (Sioris et al., 2015). In this companion paper, a second phenomenon is 18 identified that contributes secondarily to the variability of UTWV: volcanic emissions. The role 19 of volcanic emissions relative to other dynamical and thermodynamic processes in this region on 20 monthly timescales is an open question which motivates this study. 21 22 Nabro is close to the equator and Puyehue at 40degS – would it not be much more intuitive to 23 first look at water vapour profiles close to the eruptions? 24 25 We did not first look at water vapour profiles close to the eruptions since we did not know that volcanoes perturbed upper tropospheric water vapour on monthly timescales until we compiled 26 our monthly anomaly time series versus altitude (at high latitudes) and then tried to understand 27 the processes responsible for this variance. Similarly, in the companion paper, we did not set out 28 to prove that the hypothesis that the annular modes are a dominate source of variability. These 29 hypotheses only came after having plots of monthly anomaly time series at hand. 30 We were not explicit about the timescale of interest, but the duration of the volcanic impacts to 31 UTWV was mentioned in the paper and in the abstract. Also our interest in the climatic impact, 32 33 assessed through cooling rate simulations, also involves a monthly timescale. With the rewording 34 of the introduction (in response to the previous comment), we are now stating the timescale of interest explicitly. We have also removed the reference to Murcray et al. (1981), which should 35 help avoid giving the initial impression to the reader that we were focussed on shorter timescales 36 (days). 37 38 Nevertheless, since it is very likely that Puyehue was responsible for the sudden and top-ranking positive anomaly (approximately +50%) in ACE-observed southern high-latitude UTWV in the 39 2011 austral winter (July-August), it also made sense to us to look at ACE water vapour profiles 40 close to Puyehue (thus our Fig. 5) and MAESTRO-based RH profiles as close to Puyehue in 41 42 space and time as possible (e.g. Fig. 7) to understand the phase of the water. Reviewer 1 made

We have deleted Sect. 3.2 and all of the discussion regarding volcanogenic water vapour in the

the same suggestion as Reviewer 3 and so we analyzed MLS SO₂ and H₂O data of the Nabro 1 2 plume one day after eruption (see response to Reviewer 1). In writing the original manuscript, we looked at ACE mid-latitude (30-60°N) water vapour profiles close to Nabro in July 3 4 (including early July) but there are no ACE occultation events that are spatially very close to the Nabro plume in June. We did not look at low latitudes in July 2011, and fortunately there is one 5 6 ACE occultation event (ss42439) that, by fluke, fell in the Asian monsoon region (26°N, 45°W). There is clearly reduced signal-to-noise in the two ACE-FTS spectra from this occultation event 7 8 at 13.5 and 15.6 km which led to rejection of these spectra from the operational v3.5 processing. These spectra were included in the ACE-FTS water vapour retrieval by Chris Boone in response 9 to this comment (shown below). Similarly, at 14.1 and 15.1 km, MAESTRO measures water 10 vapour with >100% uncertainty due to a overlying, unusually thick cloud and/or aerosol layer 11 with 560 nm aerosol extinction peaking at 16.2 km (see Fig. 2 below). Water vapour above 15.1 12 13 km is below MAESTRO's lower detection limit for this occultation and is likely due to the reduced signal as a result of this overlying "cloud". At 13.1 km, MAESTRO measures 92±88 14 ppmv of water vapour, which translates to a relative humidity of $48\pm46\%$. This implies, in spite 15 of the huge measurement uncertainty, that the conditions are not favourable for homogeneous 16 nucleation of ice. In the spectrum immediately below 13.1 km (at 12.2 km), MAESTRO 17 18 observes a much stronger water vapour absorption signature that is likely due to an spatial inhomogeneity between spectra measured below and above 13 km. In any case, the MAESTRO 19 water vapour retrieval does not converge below 13 km. The ACE-FTS water vapour profile (Fig. 20 3 below) is consistent with the MAESTRO observation at 13.1 km: at 12.9 km, ACE-FTS 21 measures 33±5 ppmv of water vapour, again implying unsaturated air. The thermal tropopause is 22 23 at 17.5 km or 93.9 mb, where ACE-FTS water vapour is 9.9±0.2 ppmv, clearly an anomalously high value (99th percentile) in the context of Fig. 2 of Schwartz et al. (2013). Saturated air exists 24 at the tropopause according to FTS, whereas at 16.0 km, the RH inferred from ACE-FTS water 25 26 vapour is 39±13%. The cloud+aerosol extinction peak is nearer to 16.0 km however, according 27 to ACE NIR-Imager (15.5 km) and MAESTRO (16.2 km), implying a likely contribution from

- 28 Nabro aerosols to the observed cloud/aerosols at 16 km.
- 29



Figure 2. Aerosol extinction profiles observed by ACE instruments during ss42439 at 26°N, 45°W on 1 July 2011. Both instruments are shown to illustrate the agreement on profile shape and peak height. Conclusions regarding the Ångström exponent from single profiles should be avoided, partly due to difference in the size and shape of the field-of-view given possible aerosol/cloud heterogeneity over the width of the sun at the tangent point (~20 km).



Figure 3. ACE-FTS water vapour profile in the UTLS during ss42439.

However, our main focus, as stated in the title, is on high latitudes, particularly understanding UTWV variability, so observations close to the volcanoes (in space and time) is examined to make a stronger case that the high latitude variability could be due to the eruptions.

What is your argumentation that water vapour emitted near the equator should reach the polar regions (see also comment 2)?

The reviewer's question pertains to Nabro. As discussed above, we removed these arguments
 from the paper.

The authors should explain how many profiles would be available to look at the surroundings ofthe volcanoes and why they decided to not look at them (except for one profile in Fig. 7).

For Nabro, as mentioned above, there is only one profile in the surroundings of the volcano in
the first 18 days after eruption. There are no processed ACE occultation events in June 2011 in
the southern hemisphere, following the Puyehue eruption. By July 2011, it is known that the
Puyehue volcanic plume had already circumnavigated the globe twice (Vernier et al., 2013), so

zonal (40-60°S) July 2011 median data are preferable as shown in Fig. 5 of the manuscript. For

23 Eyjafjallajökull, there are many profiles in the surroundings of the volcano, however a profile on

1 May 16th, 2010 60.1°, 6.7°W shows no enhancement relative to the zonal monthly median at all

2 heights below the hygropause (~10.5 km) but it is 11 days after the second major eruption

3 (Gudmundsson et al., 2012). In April 2010, after the initial major Eyjafjallajökull eruption, ACE

4 observations are in the southern tropics and high latitudes. So there are no observations in the

5 surroundings of Eyjafjallajökull within 10 days of one of its two major eruptions in spring 2011.

6
2.) General: It is not trivial for water vapour emitted in the tropical/midlatitude troposphere to
8 reach the polar upper troposphere. As long as the air is not saturated transport is along isentropes
9 which slope upward towards the pole. Therefore air parcels moving poleward from Nabro or
10 Puyehue are expected to experience adiabatic cooling, leading to cloud formation and rainout.
11 Since I assume that the emitted air from the volcanoes is humid, it requires only a minor lifting

12 to reach saturation and cloud/rain formation.

13

14 There are two steps required if the reviewer's comment is valid. The first one is that adiabatic

15 cooling is sufficient for cloud formation. This is probably correct based on calculations for

16 Puyehue humidity at 7.5 km assuming a 1 km rise at the dry adiabatic lapse rate. The second is

17 that cloud formation leads to significant precipitation. Cloud droplets need to grow sufficiently

18 before they begin to fall. The reviewer's comment applies only in some places at some times. In 19 fact, saturation, which is a condition for cloud formation, is rather rare in the southern high

fact, saturation, which is a condition for cloud formation, is rather rare in the southern high
 latitude upper troposphere in austral winter 2011 (see discussion at bottom of p25879 continuing

to top of p25880). Furthermore, the precipitate would like be in the form of tinv ice crystals (not

rain) which could vaporize before falling too far down given the warmer temperatures below.

23 We infer that saturation/condensation did occur in some Puyehue observations (p25880L12).

For the Nabro case, there is another assumption that the reviewer is making which appears to be

completely false: according to Bourassa et al. (2012), the isentropes slope downward toward the

26 North Pole in summer of 2011. Therefore, for Nabro, air parcels moving poleward are expected

27 to experience adiabatic heating, potentially leading to melting of ice coatings on volcanic

aerosols and a local increase in water vapour.

We deleted the Nabro section and added to the Puyehue-related discussion in Sect. 4 (provided ina reply below).

31 In other words, water vapour cannot be transported easily from the tropics

to the polar upper troposphere without being deposited at the ground via precipitation.

We agree. By deleting the Nabro section, this comment is addressed.

35 Therefore studies on the typical tropospheric residence of water estimate values of a few days

36 (e.g., Trenberth, K. E. (1998). Atmospheric moisture residence times and cycling: Implications

for rainfall rates and climate change. Climatic Change, 39(4), 667-694, and several other/more
 recent studies on this topic).

- 39
- The analysis by Trenberth (1998) is not vertically resolved. Since almost all of the water vapour

41 is below 5 km, even at polar latitudes, Trenberth (1998) effectively provided the residence time

in the lower troposphere. Thus, this reference is not very relevant. Nevertheless, Trenberth
 (1998) finds atmospheric moisture residence time of 30 days in the sub-tropics based on annual

means. 30 days is also a more reasonable residence time for the high-latitude upper troposphere 1 where, similarly to the sub-tropics, precipitation is not effective at shortening residence time, 2 particularly in winter where convection is very weak at high latitudes. The high-latitude upper 3 4 troposphere should have longer residence times than the sub-tropics because of greater vertical stability in the former region. Brasseur et al. (1998) state that the water vapour residence time at 5 the tropopause is "weeks". Freeman and Liou (1981) estimate the residence time of water vapour 6 in the upper troposphere to be ~30 days. Support for these quoted residence times could not be 7 8 found. Fortunately, Ehhalt (1973) determined the residence time versus altitude in the troposphere using tritiated water measurements. At the tropopause, his estimate was three weeks 9 based on winter and spring measurements at a mid-latitude site (Nebraska, 42°N). An additional 10 minor point is that dry removal of water vapour is expected to be less efficient at high latitudes 11 due to the greater atmospheric stability and reduced surface area of snow versus forest (Prospero 12 13 et al., 1983). Vertically-resolved moisture residence times inferred from Fig. 3 of Kennett and Toumi (2005) in the sub-tropics appear too short compared to Trenberth's 30-day estimate there. 14 15 We disregard the residence times of Kennett and Toumi (2005). The data in their Fig. 3 was not available from the authors (Toumi, priv. communication). From their Fig. 3, large portions of the 16 polar upper troposphere have a residence time of >5 days, but how much greater than 5 days 17 18 cannot be said. Assuming a 3 week residence time for UTWV (Ehhalt, 1973), a simple comparison of the 19 MAESTRO-observed and simulated exponential decay shows good agreement (Fig. 4 below) at 20 8.5 km at southern high-latitudes, and there is good consistency between MAESTRO and ACE-21 FTS. Some differences between observed and simulated decays could be due to neglected 22 23 monthly changes in wind velocity (and thus to the advection of water vapour to the southern high-latitude region). Also, in September, the temperature may be cold enough for condensation 24 25 at 8.5 km which would shorten the residence time in the upper troposphere if sedimentation 26 occurs or if the resulting ice crystals tend to remain the condensed phase during that month.

28 We now write at the end of the introduction:

27 28 29

30 Water vapour at the tropopause has a typical atmospheric residence time on the order of three

31 weeks (Ehhalt, 1973; Brasseur et al., 1998) and is mostly removed by precipitation (Junge,

32 1963). The residence time decreases to ~2 weeks at an altitude of 5 km (Ehhalt, 1973) which

- 33 limits the distance over which UTWV enhancements can be advected.
- 34
- 35



Figure 4. Water vapour median anomalies in 2011 austral winter at southern high latitudes, following the Puyehue eruption (Chile). The vertical error bar is the standard error of the 2011 monthly mean.

Your statement on p. 25879/80 "most of the water emitted ... will tend to remain in the vapour phase as it is advected to the southern high-latitude upper troposphere" is most likely wrong. I think with a simple parcel model, lifting a moist air mass to the upper troposphere, you could show that saturation would occur rather quickly.

As stated above, saturation can occur, but not necessarily precipitation and the precipitation will 12 not completely remove the volcanic water vapour. Precipitation may vaporize before reaching 13 the ground given the low ambient humidity. Ice coatings may form on aerosols as a result of 14 saturation/condensation. This ice could vaporize later while the particles are still in the upper 15 troposphere. As the plume disperses, the very high humidities rarefy, and saturation will tend to 16 occur less: the saturation will tend to be mostly in the initial eruptive phase closer to the volcano. 17 18 The extratropical upper troposphere has fast winds that help to disperse the humidity and keep RH<100%. The fast winds can be inferred from the fact that Puyehue circumnavigated the globe 19 20 twice before the start of July 2011 (Vernier et al., 2013).

Of course an alternative pathway of water vapour transport is via the stratosphere. If the volcano 1 injects water (most likely in form of ice particles) into the lower stratosphere, then this vapour 2 can "survive" much longer without being trapped by clouds and could maybe make it to the polar 3 4 regions. 5 6 We agree with the comment. However, very little material, if any, from Eyjafjallajökull reached the stratosphere. For Puyehue, according to the ACE observations, the water vapour in July 2011 7 8 at middle and high latitudes was in the upper troposphere. No observations in June are available in this region. Water could have fallen from the stratosphere as ice coatings on ash during the 9 latter part of June, but the observed UTWV enhancement at mid-latitudes could be transported to 10 southern high latitudes without necessitating entry into the stratosphere (see replies above and 11 below). For Nabro, as mentioned, the section has been deleted. 12 13 No change is made to the Eyjafjallajökull or Puyehue sections of the paper. 14 15 But the paper remains very fuzzy about which transport pathway occurred, and I find it irritating 16 that the aspect of saturation and cloud formation associated with poleward transport in the 17 18 troposphere is never mentioned. 19 We thank the reviewer for the suggestion to be explicit about the transport pathway for Puvehue. 20 We have now reworded (p25879L16) as follows: 21 22 23 The consistency of these ratio profiles between middle and high southern latitudes provides evidence of the poleward transport in the upper troposphere of water vapour emitted by the 24 25 Puyehue eruption. 26 27 As we replied above, the saturation does not necessarily imply complete removal from the 28 atmosphere or even from the upper troposphere. With the rise in altitude, the resulting ice crystals may fall, but the lapse rate that led to their condensation also means that warmer 29 temperatures exist below which can result in the rapid vaporization of these ice crystals given 30 their small size, as expected for ice crystals formed at the low specific humidity of the upper 31 troposphere. The net effect is that air is transported poleward with little change in the water 32 vapour profile due to upward sloping isentropes. 33 34 We now write in Sect. 4 where Puyehue is discussed: 35 36 37 During poleward transport, air parcels follow isentropes typically to higher altitudes. Such transport involves adiabatic cooling which can lead to saturation. However, the saturation does 38 not necessarily imply complete removal from the atmosphere or even the upper troposphere. 39 With the rise in altitude, the resulting ice crystals may fall, but they may be vaporized very 40 quickly given their small size and the warmer temperatures below with the net effect being that 41 42 air parcel transported poleward on an upward sloping isentrope may experience little change in the vertical profile of the water vapour enhancement.

- 43
- 44
- 45 For Nabro, we reiterate that the section has been deleted.
- 46

2 water vapour signals, and they seem to conclude that when the volcanic aerosol plume reaches 3 the high latitudes, that then an observed water vapour enhancement is also due to the volcanic 4 plume. Again, water vapour is rather short-lived in the troposphere and responds differently to 5 cloud formation and rainout than aerosols. Therefore I would be much more careful with linking 6 volcanic aerosol plumes to water vapour signals. 7 8 The reviewer is correct that there is a danger in concluding that when the volcanic aerosol plume 9 reaches high latitudes, then the water vapour enhancement is also due to the volcanic plume. But 10 for both Puyehue and Nabro, we did not conclude solely on this fact so we were "more careful". For both cases, we showed an enhancement in water vapour at mid latitude at a consistent 11 altitude with the enhancement at high latitudes. However, in the case of Nabro, our original 12 conclusion based on both of these facts is incorrect and the section has been deleted. Thus the 13 comment will be addressed as it pertains to Puyehue. Water vapour and aerosols do not have 14 identical lifetimes in the upper troposphere but they are very similar: aerosols have a residence 15 time there of 30 days (Prospero et al., 1983; Pruppacher and Klett, 2010) and this is not 16 surprising given that the main mechanism for their removal, namely precipitation, is the same for 17 both constituents. With each volcano, we are using aerosols as a volcanic proxy. In other words, 18 if we observed enhanced water vapour in the upper troposphere without the presence of a 19 20 volcanic aerosol layer, we would reject the notion that the water vapour enhancement was 21 volcanogenic. Conversely, we understand conversely that volcanic aerosols may exist, particularly in the stratosphere, without an accompanying water vapour enhancement. Because 22 23 aerosols appear to have a slightly longer residence time in the upper troposphere, they could be also remain there while the water vapour enhancement could have been more quickly depleted. 24 Fig. 6 of the paper illustrates the monthly zonal mean aerosol extinction in July 2011 to 25 demonstrate that a volcanic aerosol layer was "initially" present in the upper troposphere at 26 27 southern high latitudes. The temporal evolution of the Puyehue UTWV enhancement at southern high latitudes is consistent with a residence time of three weeks as illustrated above (Fig. 4). 28

3) General and in line with comment 2: the authors sometimes compare aerosol signals with

29 We now write in Sect. 3.1 (p25880L9):

1

The decrease over these winter months is consistent with the lifetime of water vapour in the upper troposphere (Ehhalt, 1973).

4) p. 25874 line 15: for most readers of ACP the volcanic explosivity index is not

33 known. Therefore mentioning the index value for one eruption (but not for the others)

and without a more general context is not useful in the abstract.

A similar comment was made by Mike Fromm so we cite Newhall and Self (1982) in the revised
 manuscript. We no longer mention VEI in the abstract.

5) p. 25875 line 4: what do you mean by "in theory"? I don't think that there is a theory about this topic.

39 The reviewer seems to have missed the references provided at p25875L4. While we chose the

- 40 phrase "in theory", it appears the authors of one of the two cited papers also uses the same
- 41 language. Consider the first and third sentences of the abstract of Glaze et al. (1997):

1 Contrary to assumptions often made in the literature, explosive volcanic eruptions are capable of

2 transporting significant amounts of water into the stratosphere. (...) A theoretical model for the

3 conservation of mass, momentum, and thermal energy of four separate components (dry air,

4 water vapor, liquid condensates and solid particles) is used to determine the extent of

5 atmospheric water redistribution.6

7 Also, from their conclusion section:8

9 The theoretical results address two important issues concerning water vapor transport: (1) the 10 extent to which volcanic eruption columns are capable of entraining water vapor at lower levels 11 and (2) whether or not volcanic columns are capable of injecting significant amounts of water 12 into the stratosphere.

14 We have deleted the paragraph containing "in theory".

16 6) p. 25875 line 15: here you mention an indirect effect: volcanic eruption -> temperature

17 change -> humidity change. What I am missing here, is a systematic summary

18 of different processes of how volcanic eruptions may influence tropospheric humidity

and on what time scales (direct emission, transport, indirect effects via temperature,

20 pathway via the stratosphere, ...).

13

15

21 This line has been deleted. Discussion of a stratospheric pathway is not very relevant to the

22 revised manuscript. One indirect effect via temperature and the related reference to Soden et al.

23 (2002) has been moved to the introduction. We add the timescale to this sentence as follows:

UTWV was observed to decrease following the Pinatubo eruption due to global cooling belowthe tropopause and did not return to normal levels for two years (Soden et al., 2002).

We also add the following sentences on a temperature-related mechanism which could enhanceUTWV following a volcanic eruption:

28 For volcanoes with an eruption height at or below tropopause, local warming by radiation-

29 absorbing volcanic aerosols such as ash can lead to local increases in water vapour. The

timescale of UTWV enhancement due to such a thermal mechanism would be controlled by

rainout and fallout of the aerosol, which is on the order of ~ 1 month (Prospero, 1983; Pruppacher and Klett, 2010) for particles of intermediate size ($\sim 0.3 \mu$ m).

The residence time of UTWV and how it limits the contribution by volcanic eruptions at lower
latitudes is now provided in the introduction as well (see above).

7) p. 25875 line 17: what do you mean by "remain in the ... data": is it persistentfeature over many years?

What is meant is that the data have been reprocessed (Hurst et al., 2011) but the feature at 24-26km remains.

- 39 This sentence has been deleted.
- 40 8) p. 25875 line 23: this sentence is very long, contains different things and is confusing.
- 41 Please try to write in a clearer way.

- 1 This sentence and the entire stratospheric discussion has been deleted.
- 2 9) General: I find it strange that the coordinates of the volcanoes are never given. This
- 3 is important information.
- 4 We agree with the reviewer and have added the coordinates of the two volcanoes in the first 5 sentence of Sect. 3.1 and what is now Sect 3.2:
- 6 The Puyehue-Cordón Caulle volcano (40.59°S, 72.12°W) erupted explosively in early June of
 7 2011.
- 8 and
- 9 Eyjafjallajökull (63.63°N, 19.62°E) began erupting on (...)
- 10 10) p. 25876 line 14: cf. comment 1): Why do you mention here only high latitudes?
- 11 See reply to General comment 1) (above). A second reason for the high-latitude focus is that the 12 ACE orbit is more suited for studying processes there as compared to low latitudes.
- 13 11) p. 25876 line 19: Bernath et al. is not in the list of references.
- 14 Thanks to the reviewer for spotting this. The reference has been added.
- 15 12) Figures 1 and 2: the caption of Fig. 1 mentions VMR (of what?).
- 17 The Fig. 1 caption now reads:
- Comparison of global median water vapour VMRs from MAESTRO (blue) and ACE-FTS(black) (...)
- What should the reader learn from Fig. 2? I was confused by the many lines, instruments, errors...please help the reader to understand what is relevant for this study.
- 22

23 What the reader should learn from Fig. 2 was already provided in eight lines beginning at

- 24 p25877L5. This is a conventional validation figure. The many lines correspond to the many 25 instruments measuring UTLS water vapour profiles. The reader can see that MAESTRO and
- ACE-FTS agree fairly well and ACE-FTS and MIPAS-IMK agree fairly well in their respective
- coincidences. The middle panel shows that the rest of the instruments have large biases that
- appear at 12 km, which is typically the tropopause. The right panel ultimately shows that
- 29 MAESTRO has lacks precision in the stratosphere (but is not biased according to the middle
- panel). Only the differences between ACE-FTS and SMR exhibit more scatter. In summary, the
 middle panel tells the reader about biases and the right panel ultimately relates more to precision
 of the correlative instrument (given the very high precision of ACE-FTS).
- 33

34 It is also irritating that only the caption of Fig. 2 mentions the vertical resolution of the data. I 35 never found this discussed in the text!

36

We have inserted the following information on the vertical resolution of the MAESTRO water vapour profiles at P25877L3:

- 1 The water vapour profiles have ~1 km vertical resolution (Sioris et al., 2010).
- 2 and for ACE-FTS at P25877L22:
- 3 ACE-FTS gridded version 3.5 water vapour profiles are used in the study (Boone et al., 2013)
- 4 and are assumed to have 3 km vertical resolution.
- 5 The fourth sentence of the Fig. 2 caption now reads:
- 6 The profiles from the instrument with the coarser vertical resolution are smoothed to account for7 the difference in resolution between ACE-FTS and the correlative instrument.

8 14) p. 25878 line 8: I am not sure that your course analysis of the tropopause height is relevant.

- 10 We are not sure what is meant by "course analysis", but the tropopause definition must be
- 11 provided to the readers. Presumably, the reviewer would like finer vertical resolution of the
- 12 tropopause height ("course" -> coarse). Tropopause information is used particularly for the
- 13 Eyjafjallajökull case study where the water vapour enhancement extended up to the local
- tropopause and is relevant in light of longer residence times for water vapour in the stratosphere.
- 15 The tropopause height information comes from the GEM model which has comparable vertical
- 16 resolution to MAESTRO. It is a virtue that MAESTRO and GEM vertical resolution is very
- similar. MAESTRO and ACE-FTS are both capable of measuring temperature profiles but there
- is not an operational temperature profile product for either instrument at 10 km.
- 19 No change is made to the manuscript.
- Also Fig. 12 does not contain very interesting information. I think it would be sufficient to
 mention that the tropopause height varies between X and Y km.
- Fig. 12 has been deleted as has the entire section on Nabro.
- 23 15) p. 25878 lines 13: I don't understand this paragraph. "20 observations per altitude bin per
- 24 month": is this at a particular point or somewhere in the 60-90deg latitude band? In case of the
- latter, then I doubt that 20 observations are enough to obtain representative monthly mean, high latitude averaged profiles.
- 27
- The reviewer is correct. A circle around the Earth at a constant 60° latitude has a circumference
- of 20000 km. In order to cover all longitudes, given the spatial correlation length of water vapour at the tropopause of 400 km (Offermann et al., 2002) would require a minimum of 50
- 31 observations.
- 32 Of relevance, in 2011, there are 111, 65, 70 successfully retrieved MAESTRO water vapour
- 33 profiles for July, August, and September, respectively, at southern high latitudes to study the
- impact of the Puyehue eruption. For Eyjafjallajökull, there are 132 profiles at northern high
- 35 latitudes in May 2010. The number of ACE-FTS profiles in any given month always exceeds the
- number of MAESTRO profiles. The climatologies from each instrument are based on ~1000
- profiles since there are typically 9 populated years for each calendar month. The number of
- 38 profiles used has been added to each caption (see below).
- 39
- 40 To be clearer, we now write:

The monthly climatology, used to deseasonalize the time series, is generated by averaging the
monthly medians over the populated years, with a minimum sample size of 20 observations per
altitude bin in each individual month.

5

1

6 This section now ends with the following statement on sample sizes:

7 For the case studies presented next, there are at least 65 profiles measured by MAESTRO and by

8 ACE-FTS for each month in the July-September 2011 period at southern high latitudes

9 (Puyehue-Cordón Caulle) and for May 2010 at northern high latitudes (Eyjafjallajökull).

10 16) Figure 4 is an important figure, but I am not sure that it is consistent with Fig. 3.

11 Figure 3 shows an enormous peak in spring 2007 at 7.5 and 8.5 km, but this is not

seen in Fig. 4, which I find very irritating. Since the scale in Fig. 3 is a log-scale, this

13 peak should lead to a very prominent anomaly in Fig. 4(?).

14 The two figures are entirely consistent. Figure 3 of the manuscript shows absolute quantities

15 (monthly mean water vapour mixing ratios). Figure 4 shows relative anomalies so the large

VMRs in January (austral summer) that occur annually have been deseasonalized. Thus Figure 4

shows interannual variability only (which is true for any such figure showing relative anomalies).

18 We agree that Fig. 4 is important because it shows that at southern high latitudes, the upper

19 troposphere has low interannual variability even sampled at a monthly timescale (standard

20 deviation of 20%). This low interannual variability allows for a ~50% change in UTWV due to a

volcanic eruption such as Puyehue to stand out very clearly.

22 17) Section 3.1: I found it very difficult to understand the presentation and discussion of the

23 results in this section (which is the core part of the paper). The discussion jumps from high

24 latitudes (60-90S) to the band from 40-60S, from aerosols to water vapour, from a single profile

25 (Fig. 7) to monthly means, from VMR to relative humidity ... this really did not help to

understand the story and to find the story convincing. Please help the reader much better tofollow your line of thoughts.

29 We have added the following sentence to help guide the reader at P25879L11:

3031 To connect the clearly enhanced UTWV at southern high latitudes to the eruption of Puyehue-

Cordón Caulle (Puyehue hereafter), UTWV profiles in the 40-60°S band, which contains the
 latitude of this volcano, were contrasted between July 2011 and July 2012 (a normal July).

The reason to discuss the aerosol extinction profiles at mid and high latitudes is that aerosols serve as a volcanic proxy. In July at southern high-latitudes, it is difficult to imagine anything other than a volcano producing a widespread layer near the tropopause (as evidenced by the nearly equal median and mean extinctions in Fig. 6 of the manuscript). Because of the generally low relative humidity in the upper troposphere in July (austral winter), cirrus would not be omnipresent and would have much larger differences between monthly median and mean

extinction as is seen for the polar stratospheric clouds at ~20 km. Furthermore, the aerosol
extinction peak height at high latitudes is similar to the mid-latitude peak height (~9 km). The

43 reader is already provided with the purpose of Fig. 6 (p25879L24):

44

"...corroborates the volcanic origin of the water vapour enhancement." 1 2 We have removed the single mid-latitude RH profile from 01 July 2011 and replaced it with a 3 median RH profile for the 40-60°S latitude band in early July 2011. We appreciate the reviewer's 4 suggestion to do this. 5 6 RH is needed to determine saturation. We do not feel that the RH should be used throughout the 7 paper however since it is not the retrieved quantity from the ACE instruments (i.e. depends on 8 GEM temperature and pressure) and can reflect temperature changes as well as water vapour 9 changes. Water vapour relative anomalies are equally useful for manifesting the sudden changes 10 in UTWV arising from volcanic eruptions. 11 12 13 To justify the use of RH, we now write at p25880L2: 14 15 RH profiles (Fig. 7) are used to emphasize that most of the water emitted from the volcanic eruption will tend to remain in the vapour phase as it resides in the southern high-latitude upper 16 17 troposphere. 18 18) p. 25880 line 18: I don't understand why there is this sentence about cooling rates 19 at the surface in the paper - also the appendix does not help to understand what has 20 been done and why. 21 22 One of the reasons for studying UTWV is that it is effective at trapping longwave radiation, 23 which can lead to warmer temperatures at the surface. In the second sentence of the introduction 24 25 in the original manuscript, we state that water vapour is effective at trapping infrared radiation, particularly when it is located near the tropopause. 26 27 28 We have now made it the first sentence of the paper. 29 The paper shows that volcanic emissions can increase UTWV significantly for a period of a 30 month or two. But, we wanted to take this one step further and address the obvious climatic 31 question of whether the surface temperature would be affected by volcanic emissions of water 32 33 vapour on such a timescale. 34 Also, we have added a sentence to the introduction to help clarify why the Antarctic oscillation 35 would be included as a basis function in the multiple linear regression discussed in the appendix: 36 37 The variability of upper tropospheric water vapour (UTWV) at high latitudes is dominated by 38 dynamics (Sioris et al., 2015). 39 40 In order to understand why the cooling rate differences were simulated, we now start the 41 42 appendix with: 43 In order to investigate the impact on volcanic UTWV enhancements on surface temperature, (..) 44 45

46 We also added a final sentence to the appendix to clarify the approach:

- 2 The use of a multiple linear regression adjusts for a minor contribution by the Antarctic3 oscillation to the July 2011 UTWV enhancement.
- 5 19) General: I find the quality of the figures rather low. For instance, there are often no
- 6 axis ticks and therefore it is not clear, e.g., in Fig. 3 where 20, 30, ... ppm are. Also in
- 7 Fig. 3 some vertical lines would help a lot to attribute the values to a particular month.
- 8 Some figure captions are specific about the region, others are not. I think every figure
- 9 caption showing a profile should indicate how many profiles have been averaged to
- 10 produce the profile shown.

- 11 Ticks have been added to both axes of all figures, except for the x-axis of Fig. 3 for which
- 12 vertical gridlines separate adjacent months. Every second available month is labelled (January,
- 13 April, July, September) so labels are not present for March, May, August, and November. In
- 14 addition to the vertical gridlines, markers have been added to the four curves (i.e. altitudes) to 15 make the months easier to distinguish.
- 15 make the months easier to distinguish.
- 16 For the Fig. 2 caption, we now write:
- 17 (...) Number of coincidences globally (...)
- 18 Every other figure caption was specific about the region.
- 19 For each figure containing a vertical profile, we have added the number of profiles as follows:
- Figure 1. Comparison of global median water vapour VMRs from MAESTRO (blue) and ACE FTS (black) (N=15000).
- Figure 5. Enhancement factor for water vapour mixing ratio in July 2011 in the 40-60°S band (July 1-July 12, N=78) and the 60-66°S band (July 13-July 31, N=181) (...)
- Figure 6. ACE-Imager median and average near-infrared (NIR, 1.02 μm) aerosol extinction
 profiles for July 2011 at southern high latitudes (N=163).
- 26 Figure 7. Relative humidity for July 2011 (40-60°S, N=52) and (60-66°S, N=111) and
- climatology (60-66°S, July for every year, except 2011 between 6.5 and 9.5 km, N=865) (...)
- Figure 8. Southern high-latitude (60-90°S) monthly median water vapour profiles in July for
- 29 different years, MAESTRO: 2004-2012, ACE-FTS: 2010 (N=169) and 2011 (N=176). A
- logarithmic scale is used for the x-axis. The number of July profiles (60-90°S) for MAESTRO is
 96 per year on average.
- 32 Figure 9. (...) The uncertainty accounts for the interannual standard deviation for May (2005-
- 2012) and the relative standard error of individual profiles from the month of May 2010,
- combined in quadrature (N = 132, 178 for MAESTRO and ACE-FTS, respectively).
- Figure 10. Median and average aerosol extinction observed by MAESTRO at 560 nm in May
 2010 at northern high latitudes (N=167).
- 20) P. 25882: here I am completely lost; why do you discuss here data quality issues?
- 38 This discussion has been deleted.
- 39 40

1 References

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1 2	Water vapour variability in the high-latitude upper troposphere: 2. Impact of volcanic emissions
3	C. E. Sioris ¹ , ² , J. Zou³Zou² , C. T. McElroy²McElroy¹ , C. D. Boone⁴Boone³ , P. E.
4	Sheese ³ ,Sheese ² , and P. F. Bernath ^{4,5} Bernath ^{3,4}
5 6	[1] (Department of Physics and Engineering Physics, University of Saskatchewan, 116 Science Place, Saskatoon, SK, Canada, S7N 5E2)
7 8	[2[1] {Department of Earth and Space Science and Engineering, York University, Toronto, Canada, 4700 Keele St., Toronto, ON, Canada, M3J 1P3}
9 10	[3[2] {Department of Physics, University of Toronto, 60 St. George. St., Toronto, ON, Canada,
11	M5S 1A7}
12 13 14	[4 <u>3</u>] {Department of Chemistry, University of Waterloo, 200 University Ave. W, Waterloo, ON, Canada, N2L 3G1}
15 16	[54] {Department of Chemistry & Biochemistry, Old Dominion University, 4541 Hampton Blvd., Norfolk, VA, USA, 23529}
17	Correspondence to: C. E. Sioris (csioris@cfa.harvard.edu)Sioris (csioris@sdcnlab.esse.yorku.ca)
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19	Abstract
20	The impact of volcanic eruptions on water vapour in the region of the high latitude
21	tropopauseupper troposphere is studied using deseasonalized time series based on observations
22	by the Atmospheric Chemistry Experiment (ACE) water vapour sensors, namely MAESTRO
23	(Measurements of Aerosol Extinction in the Stratosphere and Troposphere Retrieved by
24	Occultation) and the Fourier Transform Spectrometer (ACE-FTS). The threetwo eruptions with
25	the greatest impact on the high latitude upper troposphere during the time frame of this satellite-
26	based remote sensing mission are chosen. The Puyehue-Cordón Caulle volcanic eruption in June
27	2011 was the most explosive eruption in the past 24 years and resulted in an observed (50 ± 12)%
28	increase in water vapour in the southern high-latitude upper troposphere in July 2011 that
29	persisted into September 2011. A pair of northern hemisphere volcanoes, namely Eyjafjallajökull
30	and Nabro, erupted in 2010 and 2011 respectively, increasing water vapour in the upper
31	troposphere at northern high latitudes significantly for a period of $\sim \frac{3 \text{ months following each}}{3 \text{ months following each}}$
32	eruption. Both had a volcanic explosivity index of 4. Nabro led to a statistically significant
22	

33 increase of ~1 ppm in lower stratospheric (13.5-15.5 km) water vapour at northern high-latitudes

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(60-90°N) in September 2011, when the brunt of its plume arrived in the Arctic.month. These
 findings imply that volcanogenic steam emitted into or transported to the high-latitude, upper
 troposphere during volcanic eruptions must be taken into account to properly determine the
 magnitude of the local trend in water vapour over the last decade.

5

6 1 Introduction

7 Water vapour in the tropopause region is particularly effective at trapping outgoing longwave radiation emitted by the surface (Solomon et al., 2010). Currently, trends in UTWV are not 8 9 known for high latitudes (Hartmann et al., 2013). The first step toward accurate trends is to 10 improve our understanding of UTWV variability at high latitudes. The variability of upper tropospheric water vapour (UTWV) at high latitudes is dominated by dynamics (Sioris et al., 11 2015). Water vapour is the most abundant volcanic gas, comprising over 80% by volume (Pinto 12 et al., 1989). In this companion paper, a second phenomenon is identified that contributes 13 14 secondarily to the variability of UTWV: volcanic emissions. The role of volcanic emissions relative to other dynamical and thermodynamic processes in this region on monthly timescales is 15 an open question which motivates this study. Water vapour is the most abundant volcanic gas, 16 17 comprising over 80% by volume (Pinto et al., 1989). UTWV was observed to decrease following 18 the Pinatubo eruption due to global cooling below the tropopause and did not return to normal levels for two years (Soden et al., 2002). For volcanoes with an eruption height at or below 19 20 tropopause, local warming by radiation-absorbing volcanic aerosols such as ash can lead to local 21 increases in water vapour. The timescale of UTWV enhancement due to such a thermal mechanism would be controlled by rainout and fallout of the aerosol, which is on the order of ~ 1 22 month (Prospero, 1983; Pruppacher and Klett, 2010) for particles of intermediate size (~0.3 µm). 23 24 Water vapour at the tropopause has a typical atmospheric residence time on the order of three weeks (Ehhalt, 1973; Brasseur et al., 1998) and is mostly removed by precipitation (Junge, 25 1963). The residence time decreases to ~2 weeks at an altitude of 5 km (Ehhalt, 1973) which 26 limits the distance over which UTWV enhancements can be advected. 27 TwoWater vapour in the tropopause region is particularly effective at trapping outgoing 28 longwave radiation emitted by the surface (Solomon et al., 2010). Steam emitted by volcanic 29

30 eruptions can have a lasting climatic impact when the water vapour reaches the stratosphere.

Enhanced stratospheric water vapour (up to 64 ppmv) was observed using a frost-point 1 hygrometer in the plume originating from the 18 May 1980 eruption of Mount St. Helens 2 3 (Murcray et al., 1981) near an altitude of 20 km four days later. Since then, there has been little evidence of large or long lived stratospheric water vapour enhancements, although in theory (e.g. 4 5 Glaze et al., 1997; Arfeuille, 2012), moderate enhancements are possible, particularly for tropical eruptions where entrainment of tropospheric moisture adds to the contribution from the 6 magmatic water. Stratospheric Aerosol and Gas Experiment (SAGE) II enhancements following 7 the eruption of Mount Pinatubo may be artificial given that when Halogen Occultation 8 9 Experiment (HALOE) began observing in late 1991, months after eruption, SAGE II continued to observe water vapour enhancements while HALOE (Fueglistaler et al., 2013) and UARS/MLS 10 (Elson et al., 1996) were not observing enhanced stratospheric water vapour. However, both 11 Stenke and Grewe (2005) and Joshi and Shine (2003) noted a short-lived increase in 12 13 stratospheric water vapour (of >1 ppm) observed by frostpoint hygrometers over Boulder, 14 Colorado in early 2002 and considered it to be a consequence of increased water vapour at the tropical tropopause due to local warming by Pinatubo aerosols (Considine et al., 2001). These 15 anomalous water vapour enhancements in early 1992 remain in the updated Boulder data (Hurst 16 17 et al., 2011), specifically at 2-26 km. Upper tropospheric water vapour (UTWV) was observed to decrease following the Pinatubo eruption due to global cooling below the tropopause (Soden et 18 19 al., 2002). Joshi and Shine (2003) and Stenke and Grewe (2005) also related < 1 ppm enhancements of stratospheric water vapour measured by frost point hygrometers in 1982 to the 20 21 eruption of El Chichón. The largest eruption of the past two centuries is Tambora in 1815, whose volcanic explosivity index (VEI) was 7, compared to a VEI of 6 for the 1991 Mount Pinatubo 22 eruption and is estimated to have more than doubled stratospheric water vapour (Glaze et al., 23 1997, and reference therein) at least initially. Water vapour is consumed by reaction with SO₃-in 24 25 the final step of sulphuric acid formation and also condenses on the resulting sulphuric acid particles so that local humidity can decrease even for larger injections of water into the 26 stratosphere such as produced by Toba 70000 years ago if the eruption is sulphur-rich (Bekki et 27 al., 1996). Water can enter the stratosphere as ice coatings on volcanic ash (e.g. Pieri et al., 28 29 2002). With the typically low relative humidity of the stratosphere, this ice can readily vaporize

30 before the particles fall out of the stratosphere.

1	Three recent volcanic eruptions which produced the most obvious upper tropospheric water
2	vapour enhancements: at high latitudes, namely Puyehue Cordón Caulle (June 2011), Nabro
3	(June 2011),) and Eyjafjallajökull (April 2010)), are studied here. Nabro is also shown to cause a
4	significant increase in lower stratospheric water vapour at northern high latitudes, albeit for a
5	short period (on the order of a few months), consistent with the timescale over which
6	extratropical lower stratospheric water vapour remains above the tropopause (Wilcox-et al.,
7	2012). While the climatic impact of enhanced water vapour due to the Puychue cruption is
8	shown to be minor, particularly given the short period of this voleanic enhancement, such
9	increases are relevant for UTWV trend studies, particularly if an eruption occurs near the start or
10	end of the period under consideration. using satellite-based observations. Currently, trends in
11	UTWV are not known for high latitudes (Hartmann et al., 2013). However, the main focus of this
12	work is on improving our understanding of UTWV variability at high latitudes and the role of

13 volcanic emissions relative to other dynamical and thermodynamic processes in this region (see

14 companion paper: Sioris et al., 2015).

15 2 Methods

SCISAT was launched in 2003 (Bernath et al., 2005) and the Atmospheric Chemistry 16 Experiment (ACE) datasets begin in February 2004. The satellite bears two limb sounders 17 18 measuring water vapour that both rely on the solar occultation technique: Measurements of 19 Aerosol Extinction in the Stratosphere and Troposphere Retrieved by Occultation (MAESTRO). McElroy et al., 2007) and the Fourier Transform Spectrometer (ACE-FTS) as well as an Imager 20 (Bernath et al., 2005) which provides aerosol extinction measurements (e.g. Vanhellemont et al., 21 22 2008) that can be directly compared with those retrieved from MAESTRO observations. MAESTRO is currently the only satellite instrument capable of simultaneously measuring 23 24 vertical profiles of both water vapour and extinction by fine aerosols (Sioris et al., 2010b) down 25 to the mid-troposphere. The MAESTRO water vapour retrieval relies on the 940 nm absorption 26 band and is described by Sioris et al. (2010a) and updated recently (Sioris et al., 2015), The 27 water vapour profiles have ~1 km vertical resolution (Sioris et al., 2010). Figures 1-2 present the 28 validation of MAESTRO water vapour. MAESTRO is seen to have less scatter than ACE-FTS 29 below 6.5 km. Between 6.5 and 19.5 km, the median of the relative differences between

30 MAESTRO and ACE-FTS of their individual collocated profiles is < 20%, which is also true

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1 only for MIPAS IMK data (Stiller et al., 2012) considering the other UTLS water vapour data

2 products compared in Fig. 2. However, due to the relatively large noise in the MAESTRO lower

3 stratospheric water vapour data (Fig. 2), the scatter in the relative differences between individual

4 coincident ACE-FTS and MAESTRO profiles of is on the order of ~35%, whereas those

between ACE-FTS and other atmospheric sounders are typically on the order of ~10% in this
region.

7 Sioris et al. (2010a) found a weak sensitivity of the water vapour retrieval to significant

8 perturbations in aerosol extinction. As discussed in Sioris et al. (2010a), the weaker sensitivity of

9 MAESTRO water vapour to aerosol extinction relative to other solar occultation instruments

10 which have used this absorption band, namely Polar Ozone and Aerosol Measurement (POAM)

11 III and SAGE II, is due to the availability of 'off' wavelengths (i.e. with minimal absorption by

12 water vapour) on both sides of the water vapour band, which neither of these other instruments

13 incorporated into their channel selection. This issue is also true for SAGE III (Thomason et al.,

14 2010) with neighbouring channels at 869 and 1021 nm, but to a lesser extent than for SAGE II.

15 ACE-FTS gridded version 3.5 datawater vapour profiles are used in the study (Boone et al.,

16 2013) and are assumed to have 3 km vertical resolution. This dataset has been validated as

17 discussed in the companion paper. Over the microwindows used to retrieve water vapour from

18 ACE-FTS spectra₇ (Boone et al., 2005), absorption by this trace gas is completely uncorrelated

19 with the spectrally smooth aerosol extinction signature. The insensitivity to aerosol extinction of

20 water vapour retrieved from high-resolution solar occultation spectra using microwindows is

21 well known (e.g. Rinsland et al., 1994; Michelsen et al., 2002; Steele et al., 2006; Uemera et al.,

22 2005). The ACE FTS algorithm uses this microwindow technique and usesuse of a slope term in

each microwindow accounts for the smooth aerosol extinction (Boone et al., 2005). Over each

24 microwindow used to retrieve water vapour, no higher order baseline terms are necessary. The

25 complete insensitivity to aerosol extinction is an advantage of the microwindow technique

26 relative to the band-integrated approach used in the MAESTRO water vapour retrieval. This

27 advantage is possible due to the high spectral resolution of ACE-FTS which assists in separating

the continuum level, which is monotonic over a microwindow, from the deep absorption lines

29 due to light, gas phase molecules species such as $H_2\Theta$ water vapour.

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1	The monthly tropopause height is defined by the lower of the lowest local minimum above 5 km	
2	or the lowest height above 5 km at which the lapse rate is < 2 K/km in monthly median	
3	temperatures from the Global Environmental Multiscale (GEM) regional weather forecast model	
4	(Laroche et al., 1999). Further details are given in the companion paper.	
5	To obtain a water vapour relative anomaly time series for the UTLS, the method follows that of is	
6	described by Sioris et al. (2015). The monthly climatology, used to deseasonalize the time series,	
7	is generated by averaging the monthly medians over the populated years, with a minimum	
8	sample size of 20 observations per altitude bin perin each individual month. Between 5.5 and	
9	19.5 km using 1 km vertical bins, climatological profiles are obtained for all calendar months	
10	except April, June, August, and December at northern high latitudes (60-90°N) and all months	
11	except February, June, October, and December at southern high-latitudes (60-90°S), as ACE	
12	does not sample these regions in these months. For the case studies presented next, there are at	
13	least 65 profiles measured by MAESTRO and by ACE-FTS for each month in the July-	
14	September 2011 period at southern high latitudes (Puyehue-Cordón Caulle) and for May 2010 at	
15	northern high latitudes (Eyjafjallajökull).	
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- 1 observations occur from early July to austral spring equinox covering latitudes from 60 to 81°S
- 2 with a two day absence in late August, indicating the good coverage of southern high latitudes
- 3 <u>by ACE in this season.</u> Note that the spatiotemporal sampling repeats annually for ACE as
- 4 illustrated by Randel et al. (2012). The typical 'stratospheric' monthly zonal mean values (<10
- ppm) that annually appear in September at 7.5 and 8.5 km did not appear in September 2011.
 (Fig. 3).
- 7 To connect the clearly enhanced UTWV at southern high latitudes to the eruption of Puyehue-
- 8 Cordón Caulle (Puyehue hereafter), UTWV profiles in the 40-60°S band, which contains the
- 9 latitude of this volcano, were contrasted between July 2011 and July 2012 (a normal July).
- 10 Figure 5 shows a statistically significant increase in <u>zonal median</u> UTWV in the 40-60°S latitude
- 11 band as well for July 2011 relative to a normal year (July 2012), and no significant increase
- 12 above 10 km. ACE samples the 40-60°S band in the first 12 days of the month and then samples
- 13 the $60-90^{\circ}$ S band (actually $60-66^{\circ}$ S) for the remainder of the month. The large increase in water
- 14 vapour at 8 km <u>in July 2011</u> is present in both latitude bands. The consistency of these ratio
- 15 profiles between middle and high southern latitudes provides evidence of the poleward transport
- 16 <u>in the upper troposphere</u> of UTWV water vapour emitted by the powerful Puyehue eruption (VEI
- 17 of 5) of the Puyehue-Cordón Caulle volcano
- 18 (http://www.volcano.si.edu/volcano.cfm?vn=357150)..
- 19 The anomalous, sharp peak in monthly median aerosol extinction in the southern high-latitude
- 20 upper troposphere observed by Measurements of Aerosol Extinction in the Stratosphere and
- 21 Troposphere Retrieved by Occultation (MAESTRO, McElroy et al., 2007) MAESTRO (not
- 22 shown) and ACE-Imager (Fig. 6) confirms Puyehue aerosol observations by other satellite
- 23 instruments (Vernier et al., 2013; Theys et al., 2014) and corroborates the volcanic origin of the
- 24 water vapour enhancement. The median and the mean aerosol extinction in the upper troposphere
- are nearly equal because the Puyehue aerosol layer has spread across all longitudes by July 2011
- 26 (Vernier et al., 2013). The southern high latitude upper troposphere can be quite cold in austral
- 27 winter and local condensation is known to occur (Randel et al., 2012). While there is a local
- 28 maximum in relative humidity versus altitude in the southern high-latitude upper troposphere in
- 29 July 2011, the monthly median relative humidity is only ~50% based on MAESTRO water
- 30 vapour and co-located GEM model analysis temperatures (Laroche et al., 1999),

1	meaningHowever, the widespread layer in Fig. 6 is unlikely to be due to homogeneously
2	nucleated cirrus given that the monthly median relative humidity (RH) in July 2011 is only
3	~60% at the peak (Fig. 7). This RH peak is present in the July climatology (Fig. 7) but it is much
4	more subtle, closer to the tropopause and the RH at its peak is typically half (i.e. 30%) of the July
5	2011 value. RH profiles (Fig. 7) are used to emphasize that most of the water emitted from the
6	volcanic eruption will tend to remain in the vapour phase as it is advected toresides in the
7	southern high-latitude upper troposphere (see Fig. 7). Furthermore,. At southern mid latitudes
8	(40-60°S), the earliest available MAESTRO observations of the volcanic plume by MAESTRO
9	and ACE-Imager (at 42.3°S, 70.7°W on 1(i.e. early July 2011) indicate a fine aerosol plume
10	peaking at 98.5 km (spanning 8.5-10.5 km) with relativenot shown). Relative humidity in the 40-
11	60°S band obtained using MAESTRO water vapour peakingpeaks at 6741±14% at 8.75 km (Fig.
12	7), establishing that the upper troposphere in this mid-latitude band was not saturated less than 4
13	weeksone month after the eruption. Both the mid-latitude and high-latitude RH profiles in July
14	2011 peak at 8.5 km with slightly higher relative humidity at high latitudes where the volcanic
15	UTWV enhancement encountered cooler ambient air at altitudes between 7.5 and 9.5 km.
16	Considering both the ACE-FTS (Bernath et al., 2005) and MAESTRO measurements, the largest
17	relative volcanic enhancements in water vapour in July 2011 occur at 7.5-9.5 km in July 2011,
18	where a doubling occurs is observed relative to normal mixing ratios for that month (see Fig. 8).
19	By August 2011, the relative anomaly remains of similar magnitude throughout the upper
20	troposphere, and is statistically significant (1 σ) at 7.5-8.5 km (seen by both instruments) and
21	September is enhanced slightly, particularly at 7.5 km.), whereas in September 2011, the UTWV
22	enhancement is statistically insignificant. The decrease over these winter months is consistent
23	with the lifetime of water vapour in the upper troposphere (Ehhalt, 1973). In July 2011, relative
24	humidity of 100% with respect to ice (see Murray, 1967) was reached in some profile
25	observations in the southern high-latitude upper troposphere with the corresponding MAESTRO
26	aerosol extinction observations indicating a vertically thin plume of fine particles. Thus, ice-
27	coated tropospheric aerosols are inferred to be present for these cases.
28	The large enhancement in UTWV at southern high latitudes in July 2011 however does not
29	significantly change the cooling rate at the surface (see Appendix A for details of the method).

30 **3.2** Nabro

1 Nabro crupted on 13 June 2011, but the water vapour enhancement at northern high latitudes in 2 July 2011 was minor. The ACE instruments do not observe northern high latitudes in August but 3 by September 2011, enhanced in water vapour (significant relative to quadrature sum of the interannual standard deviation and the September 2011 standard error) was observed at 10.5-11.5 4 5 km by both instruments, with MAESTRO observing the peak of the enhancement at 10.5 km and ACE-FTS at 11.5 km (Fig. 9). Both instruments agreed on the magnitude of the enhancement 6 near this ~11 km peak (51±13%, Fig. 9). The brunt of the lower stratospheric aerosol 7 enhancement from Nabro arrived in the Arctic from mid-latitudes by September 2011 riding 8 9 along the 420 K isentrope and thus descending a couple of kilometres in altitude (Bourassa et al., 2012). Figure 10 shows a significant water vapour anomaly of +30% at northern mid-latitudes 10 (30-60°N) peaking sharply at 13.5 km during summer 2011, while no other altitude level shows a 11 12 significant positive anomaly. This anomaly peak height was consistent between July and September of 2011 and the anomaly decreased from 2.4 ppm to 1.8 ppm between these two 13 14 months. Individual ACE FTS observations were examined and > 10 ppm of water vapour was not found in the stratosphere in any profile observation based on thermal tropopause heights. The 15 thermal tropopause height definition was chosen for the mid-latitude data to be more 16 17 conservative about locating the water vapour enhancements in the stratosphere in contrast to the general definition used in this work (see Sect. 2). The latitudinal sampling in 2011 at mid-18 19 latitudes was similar to other years in July and September with an average sampled latitude of <u>50°N.</u> 20 21 The positive anomaly at northern high latitudes (60-90°N) at 12.5-13.5 km is the largest on 22 record at this altitude for any calendar month in this latitude band (N = 63) for both MAESTRO and ACE FTS. The monthly median tropopause height is 10.5 km in September 2011, yet Nabro 23 appeared to increase water vapour by 30-50% at 12.5 km according to both ACE sensors for that 24 month relative to their respective climatological values. Near IR ACE Imager aerosol extinction 25 26 observations indicate an aerosol layer also peaking at 13.5 km with very little variability at that altitude (Fig. 11). The low variability provides evidence that the plume had spread zonally in the 27 northern high-latitude region three months after eruption. Figure 12 illustrates the tropopause 28 height of each of the northern high latitude ACE observations in September 2011. A tropopause 29 height of 12.5 km occurs 1% of the time and never above that altitude in this month. 30

1	In the high-latitude regions for the months affected by the three eruptions studied in this work,
2	only September 2011 at northern high latitudes had biased sampling. We determined ACE-FTS
3	water vapour anomalies for September in a narrower band (60-72°N) where the sampling is more
4	uniform from year to year than for 60-90°N. The observed enhancement profile in the two
5	latitude bands are consistent within the uncertainties of the enhancement for the 60-90°N band
6	(Fig. 13). Again, two sources of uncertainty are considered at each altitude:
7	1) the interannual variability, measured by the standard deviation of the water vapour
8	volume mixing ratio (VMR) over all Septembers, and
9	2) the variability within the month of September 2011.
10	Given the consistency of ACE-FTS water vapour between the two high-latitude bands, we rely
11	on the enhancement profile over the full high latitude region (60-90°N) since it has a larger
12	sample size. According to ACE-FTS, there is an enhancement in September 2011 relative to all
13	other Septembers between 10.5 and 19.5 km. MAESTRO measurements of this enhancement are
14	consistent with the enhancement observed by ACE FTS but have larger uncertainties in the
15	lower stratosphere as expected. The enhancement observed by ACE-FTS is 0.7±0.4 ppm at 13.5
16	km and decreases steadily with altitude to 0.4±0.3 at 15.5 km. MAESTRO does not see a
17	significant enhancement above 11.5 km when 1 km vertical binning is used. However, when the
18	water vapour VMR anomaly is calculated for a 3 km bin spanning 13.0-16.0 km and the
19	uncertainties over the three 1 km bins are combined in root-sum square fashion, the anomaly is
20	1.1±0.8 ppm in this 3 km partial column. Unfortunately, the sample sizes for October and
21	November of 2011 are currently inadequate. In January 2012, both instruments measured the
22	highest water vapour VMR at 16.5 km for any January at northern high-latitudes: 4.6 ppm and
23	4.7 ppm for MAESTRO and ACE-FTS respectively. These VMRs are statistically significant
24	enhancements for both instruments (relative to 1σ of interannual variability), with ACE-FTS
25	detecting an enhancement of 0.3±0.2 ppm. This stratospheric water vapour enhancement is
26	identical to the enhancement of 0.3±0.2 ppm determined using ACE FTS data for September
27	2011 at 16.5 km (Fig. 13) and corresponds to a subtle yet statistically significant anomaly in
28	aerosol extinction (monthly mean minus monthly median MAESTRO aerosol extinction at 525
29	nm exceeds one standard error of the mean, considering the 12-30 km range in 1 km increments)
30	that spans 15.5-17.5 km, indicating a vertical correlation of the water vapour enhancement to a

1	recent vertically localized aerosol extinction enhancement. As the contribution of the volcanic
2	aerosol diminishes due to sedimentation and diffusion, the median and average come into closer
3	agreement as the aerosol extinction observations becomes more symmetrically distributed about
4	the mean as is observed at all overlying altitudes (e.g. 18-30 km). As none of the observations in
5	January 2012 at 15.5 km (N=127) had a temperature below 195 K, polar stratospheric clouds can
6	be ruled out as an alternate cause of aerosol enhancement there and tropospheric clouds could
7	also be ruled out since the highest observed tropopause was 12.5 km. In May 2012, there are
8	only significant positive anomalies of water vapour at 18.5-19.5 km observed by both
9	instruments that correspond with a statistically insignificant MAESTRO 525 nm aerosol
10	extinction enhancement. While the Nabro aerosol perturbation may have descended, there is also
11	the possibility that the water vapour enhancement at 18.5-19.5 km is not due to Nabro. As this is
12	the most recent available May, the anomaly may simply be related to increasing stratospheric
13	water vapour from CH ₄ -breakdown as ACE-FTS shows an increasing trend (2004-2012) at both
14	18.5 and 19.5 km. Thus, we conclude that evidence for enhanced lower stratospheric water
15	vapour due to Nabro is only present up until January 2012 in the ACE sensor datasets.
10	
16	<mark>3.3<u>3.2</u> — Eyjafjallajökull ↓</mark>
16 17	3.3<u>3.2</u> Eyjafjallajökull Eyjafjallajökull <u>(63.63°N, 19.62°E)</u> began erupting on 14 April 2010 below 210 m of glacial ice
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16 17 18 19 20 21 22 23 24 25 26	3.33.2 Eyjafjallajökull Eyjafjallajökull (63.63°N, 19.62°E) began erupting on 14 April 2010 below 210 m of glacial ice (Magnússon et al., 2012), reaching an altitude of 10 km (Gudmundsson et al., 2012). This was followed by a second eruption on 05 May 2010 that also reached ~10.0 km (Gudmundsson et al., 2012). ACE does not cover northern high latitudes in April, but in May 2010, MAESTRO and ACE-FTS both see statistically significant enhancements in water vapour at 8.5-9.5 km (Fig. 149). In fact, at 9.5 km, the (69±10)% anomaly in May 2010 is the largest anomaly at this altitude in any of the 63 months that sample northern high latitudes in either dataset. The stated statistical significance considers the respective interannual variability for the month of May and the respective relative standard error for May 2010 for each dataset. The monthly mean tropopause height in May 2010 is 10.5 km but some individual observations have a tropopause
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30 differences between MAESTRO 560 nm May 2010 mean and median aerosol

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extinctionsextinction profiles and the fact that both peak at 7.5 km. The ACE-Imager NIR data at 1 northern high latitudes in May 2010 confirm an aerosol layer at 7.5±0.5 km (not shown). The 2 3 Arctic oscillation would be expected to increase water vapour by < 8% at 8.5-9.5 km in May 2010 according to the regression using year-round monthly-sampled data as determined in the 4 companion paper (Sioris et al., 2015) and is thus insufficient to explain the increase. Also, 5 although dehydrated and rehydrated layers were observed in the 2010 winter (Khaykin et al., 6 7 2013), water vapour in the upper troposphere and lower stratosphere (UTLS, 5-20 km) in the northern high latitude region in March 2010 was normal according to both MAESTRO and 8 9 ACE-FTS. ACE does not sample northern high latitudes in June, but in July, significant enhancements of water vapour of 22% and 45% remained. In July 2010, enhanced UTWV is 10 observed by both instruments only at the local tropopause (11.5 km) according to), but for 11 MAESTRO and ACE FTS, respectively. This water vapour, this enhancement coincides with the 12 13 top portion of an aerosol layer that spans 7.5 to 11.5 km according to July 2010 NIR Imager 14 observations. is not statistically significant. 15 4 Discussion In the time span of 14 months (April 2010 to June 2011), three-two extratropical eruptions with 16 VEI ≥4 occurred that had a period of significant were followed by significantly enhanced UTWV 17 18 of ~3 months, leading to monthly at high latitudes in the hemisphere of the eruption. Monthly 19 median UTWV VMR increases of up to 50%.% were observed. For Eyjafjallajökull, the enhancement was not significant in July 2011, three months after the initial eruption, and 20 21 similarly for Puyehue, the period of significantly enhanced UTWV spanned two months. While 22 eachboth of these three-impacted the high-latitude, upper troposphere in the hemisphere of the eruption, twoone of the eruptions-did not occur at high latitudes. Nabro is a tropical volcano and 23 24 Puyehue, namely Puyehue, is a southern mid-latitude volcano. Enhancements Volcanic UTWV 25 enhancements in the lower stratosphere and extratropics during the high-latitude upper tropospherecold season are more readily detected in monthly zonal median data because of the 26 27 low background VMR of water vapour in these regions. When the eruption occurs during the dry 28 half of the year (late autumn to early spring), the relative perturbation to the upper troposphere is

- 29 even larger and can last longer due to reduced rainout.this region and season, owing to the lack
- 30 of deep convection. Secondly, reduced precipitation in the wintertime high latitude upper

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-	toposphere provides a residence time for the volcance enhancement on the order of the timescale
2	of the analysis, Thus, the timing and location of the Puyehue eruption were favourable for
3	detecting its water vapour enhancement. ACE-FTS and MAESTRO indicate a ~1 ppm increase
4	in stratospheric water vapour at ~14 km at northern high latitudes due to the eruption of Nabro.
5	Nabro may be a special case because the monsoon aided the cross-tropopause flux of volcanic
6	ejecta (Bourassa et al., 2012), including ash subsequently coated in ice during tropospheric
7	ascent at southern high latitudes. During poleward transport, air parcels follow isentropes
8	typically to higher altitudes. Such transport involves adiabatic cooling which can lead to
9	saturation. However, saturation does not necessarily imply complete removal from the
10	atmosphere or even the upper troposphere. With the rise in altitude, the ice crystals that form
11	may fall, but they may be vaporized very quickly given their small size and the warmer
12	temperatures below. The net effect is that an air parcel transported poleward on an upward
13	sloping isentrope may experience little change in the vertical profile of the water vapour
14	enhancement.
14 15	enhancement. Eyjafjallajökull is likely a special case since the volcano was below > 200 m of glacial ice, some
14 15 16	enhancement. Eyjafjallajökull is likely a special case since the volcano was below > 200 m of glacial ice, some of which was vaporized in the process and rose in the eruption column. Other recent eruptions
14 15 16 17	enhancement. Eyjafjallajökull is likely a special case since the volcano was below > 200 m of glacial ice, some of which was vaporized in the process and rose in the eruption column. Other recent eruptions such as Kasatochi, which generated much more SO ₂ and whose plumes went higher into the
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14 15 16 17 18 19 20 21	enhancement. Eyjafjallajökull is likely a special case since the volcano was below > 200 m of glacial ice, some of which was vaporized in the process and rose in the eruption column. Other recent eruptions such as Kasatochi, which generated much more SO2 and whose plumes went higher into the atmosphere than Nabro (and Eyjafjallajökull), were observed to have little impact on stratospheric water vapour. It is interesting to note that Eyjafjallajökull (Sears et al., 2013) and Puyehue (Pumphrey et al., 2015; Vernier et al., 2013) emitted relatively little SO2 considering their VEI values, thereby allowing less volcanic water vapour to be consumed by the reaction
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14 15 16 17 18 19 20 21 21 22 23	enhancement. Eyjafjallajökull is likely a special case since the volcano was below > 200 m of glacial ice, some of which was vaporized in the process and rose in the eruption column. Other recent eruptions such as Kasatochi, which generated much more SO ₂ and whose plumes went higher into the atmosphere than Nabro (and Eyjafjallajökull), were observed to have little impact on stratospheric water vapour. It is interesting to note that Eyjafjallajökull (Sears et al., 2013) and Puyehue (Pumphrey et al., 2015; Vernier et al., 2013) emitted relatively little SO ₂ considering their VEI values, thereby allowing less volcanic water vapour to be consumed by the reaction which converts SO ₃ to sulphuric acid and also reducing the probability of water uptake by the resulting sulphate aerosol. Volcanic emissions are known to be more variable in terms of SO ₂
14 15 16 17 18 19 20 21 22 23 23 24	enhancement.Eyjafjallajökull is likely a special case since the volcano was below > 200 m of glacial ice, some of which was vaporized in the process and rose in the eruption column. Other recent eruptions such as Kasatochi, which generated much more SO2 and whose plumes went higher into the atmosphere than Nabro (and Eyjafjallajökull), were observed to have little impact on stratospheric water vapour. It is interesting to note that Eyjafjallajökull (Sears et al., 2013) and Puyehue (Pumphrey et al., 2015; Vernier et al., 2013) emitted relatively little SO2 considering their VEI values, thereby allowing less volcanic water vapour to be consumed by the reaction which converts SO3 to sulphuric acid and also reducing the probability of water uptake by the resulting sulphate aerosol. Volcanic emissions are known to be more variable in terms of SO2 than water vapour (Pinto et al., 1989).

26 <u>5 Conclusions</u>

- 27 Due to the sporadic nature of volcanic eruptions, the UTWV variability explained by volcanic
- 28 <u>emissions at high latitudes over a decade is much less than is attributable to the annular mode of</u>
- 29 internal variability. However, this study shows that volcanic emissions can lead to UTWV

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T	increases on a monthly timescale of >50%, comparable to the U1 w v increases observed during
2	the largest annular mode negative events (Sioris et al., 2015).
3	While the climatic impact of enhanced water vapour due to the Puyehue eruption is shown to be
4	minor, particularly given the short period of this volcanic enhancement, such increases are
5	relevant for UTWV trend studies, particularly if an eruption occurs near the start or end of the
6	period under consideration.
7	Finally, MAESTRO, a solar occultation instruments, particularly those instrument operating at
8	visible and near-infrared wavelengths, havehas the unique capability among current space-borne
9	instruments to simultaneously observe vertical profiles of aerosol extinction and water vapour in
10	the UTLS to provide an understanding of the impact of volcanic emissions on the water vapour
11	budget and trends in water vapour.
12	
13	Appendix A: Cooling rate differences
14	CoolingIn order to investigate the impact on volcanic UTWV enhancements on surface
14 15	CoolingIn order to investigate the impact on volcanic UTWV enhancements on surface temperature, cooling rate vertical profiles are calculated for July 2011 using MODTRAN5.2 (e.g.
14 15 16	CoolingIn order to investigate the impact on volcanic UTWV enhancements on surface temperature, cooling rate vertical profiles are calculated for July 2011 using MODTRAN5.2 (e.g. Bernstein et al., 1996) assuming an Antarctic surface altitude of 2.5 km, <u>the</u> tropospheric
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- 3 simulations for the Puyehue eruption. We appreciate the availability of the AO and AAO indices
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Figure 2-. (left) Number of coincidences globally as a function of altitude between ACE-FTS 6 7 and various limb sounders that measured water vapour in the ACE time period. The coincidence 8 criteria are < 1 hour in time and within 250 km. (centre) Median of relative differences in water vapour versus ACE-FTS (the minuend). ACE-FTSThe profiles are assumed to have 3 kmfrom 9 the instrument with the coarser vertical resolution and the smoothing accounts are smoothed to 10 account for the finitedifference in resolution of between ACE-FTS and the correlative 11 instruments instrument. ACE-FTS has coarser vertical resolution than most of the chosen 12 13 instruments. (right) Variability of the relative differences. SAGE is the Stratospheric Aerosol and Gas Experiment. MIPAS IMK is the Michelson Interferometer for Passive Atmospheric 14 Sounding water vapour product developed at the Institut für Meteorologie und Klimaforschung 15 16 (IMK). The MIPAS water vapour product from the European Space Agency (ESA) is also illustrated. SMR is the sub-mm radiometer on Odin and Aura MLS (Microwave Limb Sounder) 17

- 18 is used.
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Figure 3. Monthly mean time series of MAESTRO water vapour mixing ratio at different heights
(indicated in legend, in km) in the southern high-latitude tropopause region. Months of February,
June, October, December are not included as ACE does not sample in this region during those

5 months. Discontinuities indicate insufficient data during the other eight calendar months. A

6 logarithmic scale is used for the y-axis.





2 Figure 4. MAESTRO relative monthly median water vapour anomalies at 7.5 and 8.5 km at

³ southern high-latitudes ($60-90^{\circ}$ S).







Figure 5. Enhancement factor for water vapour mixing ratio in July 2011 in the 40-60°S band

3 (July 1-July 12, N=78) and the 60-66°S band (July 13-July 31, N=181), relative to July 2012.

4 The error bar on the ratio profiles account for 1 standard error of the <u>MAESTRO</u> monthly mean

5 for both years, combined in quadrature.





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2 Figure 6. ACE-Imager median and average near-infrared (NIR, 1.02 μm) aerosol extinction

3 profiles for July 2011 at southern high latitudes-(N=163). The small differences between median

- 4 and average extinction near the peak indicate a widespread layer in the tropopause region. One
- 5 standard error of the monthly mean is shown as the error bar.







9 the July 2011 RH profiles only accounts account for the standard error of the monthly mean water

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vapour.

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2 Figure 8. Southern high-latitude (60-90°S) monthly median water vapour profiles in July for

- 3 different years -(, MAESTRO: 2004-2012, ACE-FTS: 2010-(N=169) and 2011 (N=176). A
- 4 logarithmic scale is used for the x-axis. <u>The number of July profiles (60-90°S) for MAESTRO is</u>
- 5 <u>96 per year on average.</u>



2 Figure 9. Water vapour relative anomaly at northern high-latitudes in September 2011 (when the

3 stratospheric aerosol optical depth enhancement due to Nabro peaked in this region). The

4 uncertainties reflect the combined natural and instrumental variability (interannual variability

5 (1σ) for September (2004-2012) added in quadrature with the relative standard error of

6 individual September 2011 observations).





2 Figure 9. Figure 10. MAESTRO water vapour anomaly at northern mid-latitudes for summer

3 2011 (average of monthly median anomalies from July and September 2011 data). The

- 4 uncertainty represents the standard deviation of the July and September anomalies (2004-2012,
- 5 $12 \le N \le 14$, depending on altitude).





2 Figure 12. Histogram of tropopause heights in individual soundings in September 2011 at

- 3 northern high latitudes.
- 4
- 5



2 Figure 13. September 2011 median water vapour absolute anomaly based on MAESTRO and

3 ACE-FTS northern high-latitude observations (60-90°N) with the uncertainties accounting for

4 the quadrature sum of the interannual September variability (2005-2012) and the standard error

5 of the individual observations for the month of September 2011 (separately for each instrument).

6 FTS60-72 indicates the anomaly profile for the same time period limiting the observations to a

7 narrower range (60-72°N) which is more uniformly sampled from year-to-year. Uncertainties are

8 missing to the left side of the profile when they exceed 100%.



Figure 14. Water vapour relative anomaly in May 2010 at northern high latitudes following the
Eyjafjallajökull eruption. The uncertainty accounts for the interannual standard deviation for
May (2005-2012) and the relative standard error of individual profiles from the month of May
102010, combined in quadrature, (N = 132, 178 for MAESTRO and ACE-FTS, respectively).





3 <u>Figure 10.</u> Median and average aerosol extinction observed by MAESTRO at 560 nm in May

- 4 2010 at northern high-latitudes-(N=167).
- 5