

Vertical structure of stratospheric water vapour trends derived from merged satellite data

Article

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1 *Vertical structure of stratospheric water vapour trends derived from merged*
2 *satellite data*

3
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7
8 **Stratospheric water vapour is a powerful greenhouse gas and knowledge of its**
9 **long-term behaviour is crucial to understand climate change. The longest**
10 **available record from balloon observations over Boulder shows increases in**
11 **stratospheric water vapour which cannot be fully explained by observed**
12 **changes in its main drivers, tropical tropopause temperatures and methane.**
13 **Although satellite observations could help resolve the conundrum,**
14 **constructing a reliable long-term data record is challenging. Here we**
15 **introduce a new data merging approach using a chemistry-climate model**
16 **nudged to observed meteorology as a transfer function between satellite**
17 **datasets, overcoming issues arising from instrument drifts and short overlap**
18 **periods. In the lower stratosphere, the resulting water-vapour record is**
19 **extended back to 1988 and largely follows tropical tropopause temperatures.**
20 **Lower and mid-stratospheric long-term trends are negative rather than**
21 **positive, with Boulder trends shown not to be globally representative. In the**
22 **upper stratosphere, the record is extended back to 1986 and shows positive**
23 **long-term trends. The altitudinal differences in the trends are explained by**
24 **methane oxidation together with a strengthened lower-stratospheric and**
25 **weakened upper-stratospheric circulation inferred by this analysis. Our**
26 **results call into question previous estimates of surface radiative forcing based**
27 **on presumed long-term global lower-stratospheric water-vapour increases.**

28
29 Recent experiences with climate data records suggest that there is nothing like ‘the
30 ultimate climate data record’ and that different approaches to dataset construction
31 are needed to estimate the uncertainty introduced by the construction process itself.
32 For example, upper-tropospheric warming was underestimated by Microwave-
33 Sounding Unit Channel 2 temperature due to the influence of a priori information
34 and the coarse vertical resolution of the retrieval¹. More recently, it has been
35 argued² that the apparent hiatus in global-mean warming is an artefact of sampling
36 biases in the global network of surface data used to estimate global mean
37 temperature changes. Both examples illustrate the limitations of observational
38 datasets with gaps filled by statistical relationships.

39
40 An important climate data record is stratospheric water vapour, which exerts a
41 strong radiative forcing affecting temperatures both locally³ and at Earth’s
42 surface^{4,5}. Through thermal-wind balance, stratospheric temperature changes are
43 believed to affect the stratospheric circulation and, through dynamical coupling,
44 surface climate^{6,7}. Long-term changes in extratropical lower-stratospheric water
45 vapour derived from balloon measurements at Boulder (the longest available
46 record⁸) from 1980-2010 show an average increase of 1.0 ± 0.2 ppmv in the 16-26

47 km altitude range^{9,10}. About 25-30% of this increase has been attributed to methane
48 oxidation^{10,11}. The rest remains unexplained, since tropical tropopause
49 temperatures (another key driver of long-term changes¹²) exhibit trends that are
50 not significantly different from zero over this period^{13,14}. Comparison of the Boulder
51 record with HALOE satellite measurements, which exhibit essentially a zero long-
52 term trend from 1992-2005, shows discrepancies in the early 1990s¹⁵. However,
53 there is a possibility that the HALOE record suffers from aerosol contamination or
54 long-term drifts¹⁶.

55

56 The observed records of stratospheric water vapour thus present a conundrum. As a
57 result, confidence in global long-term trends is low^{17,18}. The difficulty in quantifying
58 stratospheric water-vapour trends arises from limitations of observational systems
59 in the face of strong interannual and decadal variability^{15,19,20}. There is general
60 agreement that upper-tropospheric and lower-stratospheric humidity
61 measurements from the global radiosonde network cannot be trusted²¹. Balloon-
62 borne frostpoint hygrometers are characterized by high accuracy and precision²²,
63 but their measurement records are temporally and spatially sparse. Satellite
64 instruments offer global coverage but have finite lifetimes, so different datasets
65 need to be merged into long-term records, often without much overlap. Even with
66 overlapping datasets, the merging may introduce temporal inhomogeneities since
67 aging instruments can show degradation in performance.

68

69 **New approach to merge stratospheric water vapour datasets**

70

71 We introduce a new approach to investigate long-term trends in stratospheric water
72 vapour, using timeseries from a state-of-the art chemistry-climate model nudged to
73 observed meteorology (but not water vapour) from the ERA-Interim reanalysis over
74 1980-2010 (CMAM30) as a transfer function between satellite datasets. This
75 approach exploits the extensive effort made in developing stable reanalysis
76 products, with ERA-Interim now exhibiting a much better representation of the
77 stratospheric circulation than earlier products²¹. The resulting CMAM30
78 stratospheric water vapour is expected to provide a reasonable long-term reference
79 since it includes the main known transport, mixing, microphysical (dehydration at
80 the tropical tropopause and in polar regions), and chemical processes (in particular
81 methane oxidation) affecting its distribution and long-term changes. Although the
82 model is not assumed to be correct in absolute terms, its use as a transfer function
83 allows relative biases between satellite instruments to be determined using all
84 available measurements, not just those restricted to overlap periods, thereby
85 improving the characterization of inter-instrument biases and allowing the
86 identification of potential instrumental drifts or sampling issues. Consistency
87 between model and measurements suggests that the processes controlling
88 stratospheric water vapour are sufficiently well understood to explain the long-term
89 changes, and that CMAM30 can be trusted as a transfer function, while
90 inconsistencies point out weaknesses either in the model or observations. The
91 temporal homogeneity of the water-vapour record can also be tested by examining
92 the consistency of its long-term changes with those of other variables. Using this

93 knowledge, the observational datasets can more confidently be used to create long-
94 term data records.

95
96 As an application of the approach, we merge zonal monthly-mean water-vapour
97 timeseries from seven limb-viewing satellite instruments compiled and quality
98 assessed by the SPARC Data Initiative²³ into a long-term record. *Figure 1a* shows the
99 individual satellite timeseries at 100 hPa for 20S-20N and the large discrepancies
100 between them. Relative biases to CMAM30 are calculated for each instrument
101 (*Figure 1b*), avoiding periods where the instruments have known problems (see
102 Supplementary Material). The post-2006 period is excluded from the relative-bias
103 calculation because of a known inhomogeneity in ERA-Interim lower-stratospheric
104 temperatures in late 2006 due to the introduction of GPS radio-occultation data²⁴
105 (see *Supplementary Material, Table 1*). Overall, the relative biases are seen to be well
106 defined (as shown in scatter plots in *Supplementary Figure S1*), yielding confidence
107 in the ability of CMAM30 to represent water-vapour variability, and thus in its use as
108 a transfer function between datasets. Using CMAM30 as a transfer function, each
109 instrument record is then adjusted relative to Aura-MLS (*Figure 1d*).

110
111 There is a potential pitfall in this approach in that long-term changes in the merged
112 dataset could be influenced by the long-term trend in the model. This possibility is
113 assessed by examining whether there are apparent drifts in the model-
114 measurement differences over time, or jumps between the older (SAGE II and
115 HALOE) and newer instruments following the bias correction (*Figure 1c*). For the
116 most part, the differences between these bias-corrected timeseries and CMAM30 are
117 stable in time, suggesting that there is no artificial long-term trend introduced by
118 this procedure. That the differences are stable over the SAGE II record furthermore
119 indicates very good long-term stability of these observations, despite earlier
120 concerns about a drift in the instrument's retrieval channel²⁵. This suggests that
121 SAGE II can be used to extend the satellite water-vapour record back to the mid-
122 1980s. Distinct low biases are found for HALOE during 1993-1995, which are likely
123 due to aerosol interference in the retrieval after the Mt. Pinatubo eruption, and
124 during 2003-2005, confirming previous comparisons^{16,20}. Similarly, the last year of
125 SAGE II data (2005) seems to exhibit a low bias. The earlier MIPAS data (2002-
126 2004) indicate a slight low bias with respect to the later MIPAS data (2005-2010), as
127 also found in Ref 23. The remaining fluctuations reveal mostly differences in how
128 the instruments resolve the amplitude of the seasonal cycle, likely attributable to
129 differing vertical resolutions of the observations²³. The bias-corrected timeseries
130 show a coherent evolution of tropical lower-stratospheric water vapour (*Figure 1d*),
131 with no evidence of a jump between the older and newer instruments — further
132 evidence that the procedure has not introduced any artificial long-term trend. A
133 merged satellite stratospheric water-vapour record is finally produced by
134 calculating the multi-instrument mean of all available bias-corrected datasets
135 (however excluding HALOE during 2003-2005 and SAGE II during 2005 due to their
136 identified low biases).

137
138

139 **Consistency with tropical tropopause temperatures**

140

141 In the lower stratosphere, water vapour is known to broadly follow variations in
142 tropical tropopause temperatures^{15,16,17,19,20,27,28}. The merged record is therefore
143 compared to temperature fluctuations, using deseasonalized anomalies normalized
144 by the standard deviation of the respective variable's interannual variability in
145 order to make them comparable and check their consistency (*Figure 2a*). We here
146 use the CMAM30 100 hPa temperature averaged over 15S-15N, which has been
147 shown to vary coherently with cold-point tropopause temperatures^{19,28}, and
148 emphasize again that the variability and trends of the individual datasets are
149 unaffected by the bias correction and thus not influenced by the model. The 80 hPa
150 water-vapour anomalies derived from the merged satellite record (at this level
151 representative of purely stratospheric air) strongly follow the temperature
152 fluctuations, with a correlation coefficient (R) (or variance explained) of 0.77 (59%)
153 over the full time period, which increases to 0.89 (78%) when considering only
154 2001 onwards (likely explained by the better spatio-temporal coverage provided by
155 the newer instruments, resulting in more representative zonal monthly means). The
156 consistency between the temperature and water-vapour datasets is further
157 highlighted by plotting the normalized differences (or residuals) of their anomalies
158 (*Figures 2b and c*), for which the inter-annual variability is much reduced.

159 Consistency between the merged water-vapour record and tropical tropopause
160 temperatures is also found in the extratropical lower stratosphere at 100 hPa (with
161 a lag of two months to account for transport time scales between the tropics and
162 extratropics²⁸)(*Figures 2d and e*), with a correlation coefficient (variance explained)
163 of 0.66 (43%). The normalized differences between water vapour and temperature
164 are somewhat stronger than in the tropics, due to enhanced dynamical variability
165 and its effect on tracer transport and mixing with older stratospheric air at these
166 latitudes. Dehydration in the polar vortex may also contribute²⁹. The good
167 agreement between measurements and model (*Figure 2e*) shows that these
168 additional processes are adequately represented in the model. Nevertheless, the
169 low-frequency variability of extratropical lower-stratospheric water vapour seems
170 mainly to arise from the variability of tropical tropopause temperatures.

171

172 There are four time periods that show deviations from this strong correlation: 1992-
173 1996 for the observations (but not for the model), which is presumed to be a result
174 of Mt Pinatubo aerosol affecting the water-vapour retrieval¹⁶; and 1999-2000, 2003,
175 and 2008-2009. The causes of the latter are not known but since they occurred in
176 both observations and model they are presumed to be real.

177

178 The new merged water-vapour record appears to be an improvement over a
179 previous merge of the HALOE and Aura-MLS data sets based only on the relative
180 bias during their 16 months of overlap^{16,17,30}, since the latter record shows a
181 temporal inhomogeneity in deviations from the temperature record (*Figures 2c,e*).
182 As discussed above, HALOE exhibits a low bias in the lower stratosphere during its
183 final years of operation that may adversely affect a long-term data record

184 constructed by merging HALOE with another dataset such as Aura-MLS during this
185 period. Our approach, in contrast, shows that taking the long-term behaviour of the
186 datasets into account substantially improves the consistency between the water-
187 vapour and temperature records. After the strong dip around 2001, the previous
188 merge of HALOE and Aura-MLS only partially recovers by 2010 (Refs 30,31), while
189 the merged record using CMAM30 recovers fully to pre-2001 values by 2007.
190 Calculations of surface radiative forcing from changes in lower-stratospheric water
191 vapour based on the simple merge of HALOE and Aura-MLS³⁰ may thus
192 overestimate the cooling effect on global mean surface temperatures after 2001.

193 194 195 **Comparison with in-situ observations over Boulder**

196
197 We now turn to the question of the apparent inconsistency between the long-term
198 Boulder FPH balloon and satellite datasets^{15,17}. We investigate whether the Boulder
199 trends are representative of the Northern Hemisphere mid-latitude stratosphere by
200 sub-sampling the model at the location (40N/105W) and time of the Boulder
201 measurements. *Figure 3a* shows Boulder 100 hPa water-vapour anomalies
202 extending back to 1980 together with the full and subsampled anomalies derived
203 from the model. The agreement between the merged satellite and full model
204 datasets back to 1988 provides confidence that the model exhibits a correct
205 representation of inter-annual variability and long-term changes in stratospheric
206 water vapour. The sub-sampled model fields generally correlate better with the
207 Boulder in-situ measurements than do the full model fields in terms of year-to-year
208 fluctuations, explaining the differences between the Boulder and (zonal-mean)
209 satellite observations during some years (e.g., 1988-1992, 1997-1998, and 2003-
210 2005). Nevertheless, the 100 hPa change over 1980-2010 derived from the sub-
211 sampled model fields (-0.27 ± 0.18 ppmv) disagrees with that from Boulder (0.6 ± 0.15
212 ppmv). The difference is smaller, but still statistically significant (-0.15 ± 0.22 ppmv
213 compared with 0.39 ± 0.18 ppmv), over 1988-2010 where the zonal-mean model
214 trend is consistent with that of the merged satellite record. The differences from the
215 near-global water-vapour fields (*Figure 3b*) illustrate in more detail the close
216 agreement between the sub-sampled and Boulder water-vapour records, except for
217 the three time periods highlighted in red which together lead to the differences in
218 their long-term trends. Inspection of the model's longitude-latitude distribution of
219 water-vapour changes indicates that these are not longitudinally uniform (*Figure*
220 *3c*). Positive trends are found south-west of Boulder, with the limited spatial
221 resolution of the model likely missing the full extent of the geographical structure
222 and temporal variability of this feature. Our results suggest that the water-vapour
223 trends over Boulder should not be considered representative of the global
224 stratosphere.

225 226 **Long-term stratospheric water-vapour changes**

227

228 *Figure 4a* shows the bias-corrected individual water-vapour datasets at 10 hPa in
229 the extratropics together with the model-instrument biases (*Figure 4b*). Our method
230 reveals minor discrepancies between two available SAGE II data versions (see
231 Supplementary Material). HALOE shows no apparent low bias as was identified in
232 the lower stratosphere for its last years of operation, showing that satellite
233 instrument biases and drifts can be latitude- and altitude-dependent. While the
234 model exhibits a strong low bias at this altitude, its long-term evolution and inter-
235 annual variability show very good agreement with the observations back to 1986
236 (*Figure 4c*). This level of agreement provides confidence in the ERA-Interim
237 reanalysis driving the model while at the same time highlighting once again the high
238 quality of the SAGE II data, suggesting that the satellite water-vapour record can be
239 extended back to the mid-1980s. (See *Supplementary Figure S3* for more examples.)
240

241 *Figure 5a* shows the long-term changes between the late 1980s and 2010 derived
242 from the merged satellite record throughout the stratosphere, and *Table 1* quantifies
243 the long-term changes shown in the different figures. The trends are significantly
244 positive in the upper stratosphere, while the lower and mid-stratosphere show
245 significant negative trends (in contrast to the Boulder observations). This vertical
246 structure in the long-term trends is found at all latitudes. In the tropical tropopause
247 region around 80 hPa, a negative long-term trend is identified with 70%
248 significance. On the other hand, positive changes of more than 10% are found in the
249 tropical upper troposphere. While these latter two findings need to be treated with
250 caution due to sampling limitations, they are in broad agreement with past trends
251 derived from chemistry-climate model simulations^{12,32}.
252

253 The observed water-vapour changes are now attributed to different drivers using
254 the well-established “total water” diagnostic^{33,34,16} (see *Methods* and *Supplementary*
255 *Material*). The contribution from methane entry-value changes is shown in *Figure*
256 *5b*, and is derived from tropospheric observations of methane changes together with
257 a fractional-release factor (α) (*Supplementary Figure S3*) inferred from ACE-FTS
258 stratospheric methane measurements. The contribution varies smoothly from zero
259 in the tropical lower stratosphere to approximately 3% of water vapour in the
260 upper stratosphere, the latter representing a significant fraction of the observed
261 water-vapour increase (*Figure 5a*). The contribution from water-vapour entry-value
262 changes is obtained from the merged 80 hPa record shown in *Figure 2a*, and is a
263 constant -0.14 ± 0.2 ppmv (hence not plotted). Assuming conservation of total water,
264 the difference between the sum of those two contributions and the observed change
265 can be attributed to changes in α , representing a change in stratospheric circulation,
266 whose inferred contribution to the observed water-vapour change is shown in
267 *Figure 5c*. This contribution is negative in the lower stratosphere and positive in the
268 upper stratosphere. The breakdown of the different contributions is shown in *Figure*
269 *5d* together with their uncertainties for three locations with particularly large long-
270 term water-vapour changes. The uncertainty in the inferred contribution from
271 circulation changes is dominated by the large uncertainty in the water-vapour
272 entry-value changes because of the large inter-annual variability of the latter (*Figure*

273 2a). However, the difference between the inferred contributions in the upper and
274 lower stratosphere is robust because the same water-vapour entry-value change is
275 used for both, and it is not possible to explain the observed water-vapour changes,
276 within uncertainties, without the inferred circulation changes. In the northern high-
277 latitude upper stratosphere (5 hPa, 62.5N) the long-term increase of 0.28 ppmv is
278 due in equal measure to methane increase and α increase, with an offset from the
279 decreasing water-vapour entry value. In the midlatitude lower stratosphere of both
280 hemispheres (30 hPa, 42.5N and 50 hPa, 43.5S), long term decreases of -0.28 and -
281 0.34 ppmv respectively are mainly explained by decreasing α , with an additional
282 contribution from the decreasing water-vapour entry value and a small offset from
283 the methane increase. Thus, the different drivers affect the water-vapour changes
284 differently in different regions.

285
286 Larger α corresponds to older age-of-air. To facilitate comparison with observed
287 estimates of long-term circulation changes, we determine an approximate
288 relationship between the two quantities (*Supplementary Figure S5*), and use it to
289 translate the inferred α changes into age-of-air changes (*Figure 5e*). This shows a
290 strong decrease in age-of-air in the lower stratosphere, and a weak increase in the
291 upper stratosphere, which is broadly consistent with the evidence for both
292 tendencies in long-term observations of stratospheric trace gases^{35,36,37}. An
293 increased strength of the lower-stratospheric circulation is also consistent with
294 chemistry-climate model simulations³⁸, likely exacerbated in past decades by the
295 effect of the ozone hole, which will reverse sign in the future³⁹.

296 297 **Improved knowledge of stratospheric water-vapour changes**

298
299 We have introduced a novel method to generate a long-term record of stratospheric
300 water vapour, using a chemistry-climate model nudged to observed meteorology to
301 provide a transfer function between the available satellite datasets. This approach
302 provides an improved assessment of the relative biases between instruments,
303 potential instrumental drifts, as well as possible sampling biases, compared to what
304 is possible from the observations alone, especially for instruments with no or small
305 overlap in time.

306
307 The new merged satellite water-vapour record extends back to the late 1980s and
308 shows long-term decreases in the lower and mid-stratosphere, in contrast to the
309 Boulder record which is shown not to be globally representative. Upper-
310 stratospheric water vapour instead shows a long-term increase. The contributions
311 of the two recognized drivers of water-vapour changes — the stratospheric entry
312 values of water vapour and of methane — are quantified and shown not to be
313 sufficient to explain the observed water-vapour trends, particularly the difference in
314 the trends between the upper and lower stratosphere. The discrepancy is attributed
315 to changes in the fractional-release factor between methane and water vapour,
316 which imply a strengthened lower-stratospheric circulation (reduced age-of-air)
317 and a weakened upper-stratospheric circulation (increased age-of-air), consistent

318 with other evidence. It hence appears necessary to consider long-term changes in
319 the stratospheric circulation when interpreting changes in stratospheric water
320 vapour, together with changes in methane and water-vapour entry values.

321
322 Our results show the value of using models and measurements together to
323 understand the interannual and long-term behaviour of stratospheric water vapour,
324 with the approach being applicable in principle to other trace gases. They also
325 highlight the need for independent and redundant global measurement systems
326 characterized by high long-term accuracy (and precision) to be able to quantify
327 long-term changes in stratospheric water vapour with more confidence.

328

329

330 **Acknowledgements**

331

332 We acknowledge the Canadian Space Agency for having funded the CMAM30
333 project, with additional institutional support from the Canadian Centre for Climate
334 Modelling and Analysis who provided the model code and supercomputing time. We
335 thank all national and international space agencies for making available their limb
336 satellite observations for use in the SPARC Data Initiative.

337

338

339 **Author contributions**

340

341 M.I.H. designed the methodology, performed the data analysis, and wrote the paper.
342 D.A.P. helped with the statistical analysis and together with J.F.S. devised and
343 implemented the nudged model simulations. T.G.S contributed to the interpretation
344 and writing of the text. D. H. provided processed balloon observations. J. A., L. F., B.
345 F., A. R., J. U., T. v. C., H. J. W., K. A. W., S. T. and K. W. processed and provided the
346 satellite datasets.

347

348 **Additional information**

349

350 Supplementary material is provided. Correspondence and request for materials
351 should be addressed to M.I.H.

352

353 **Competing financial interests**

354

355 The authors declare no competing financial interests.

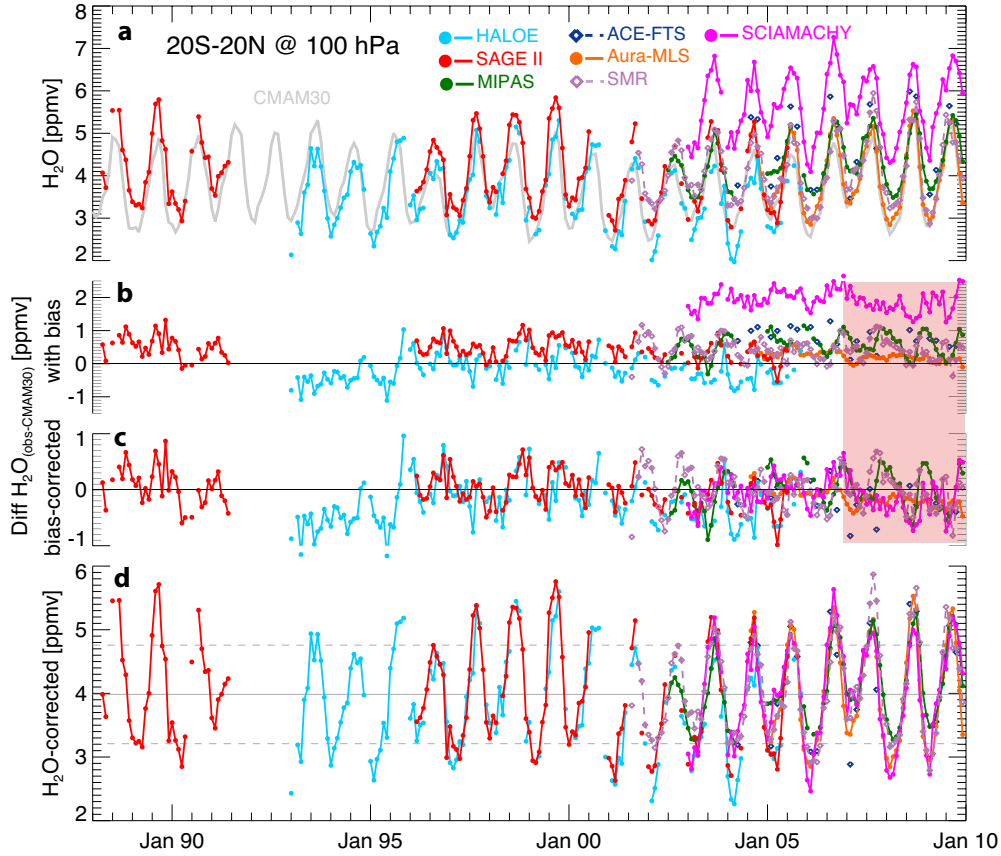
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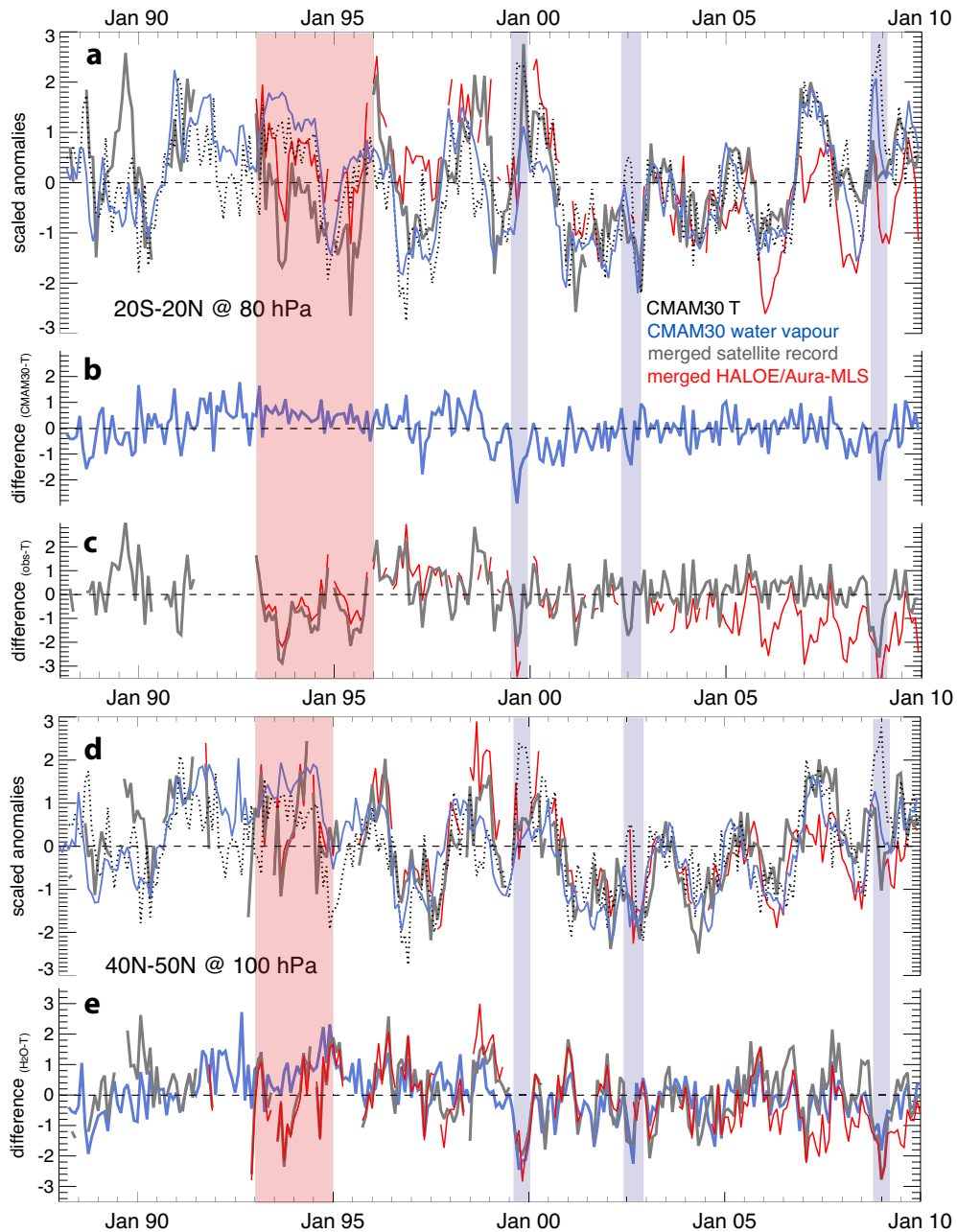
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FIGURES



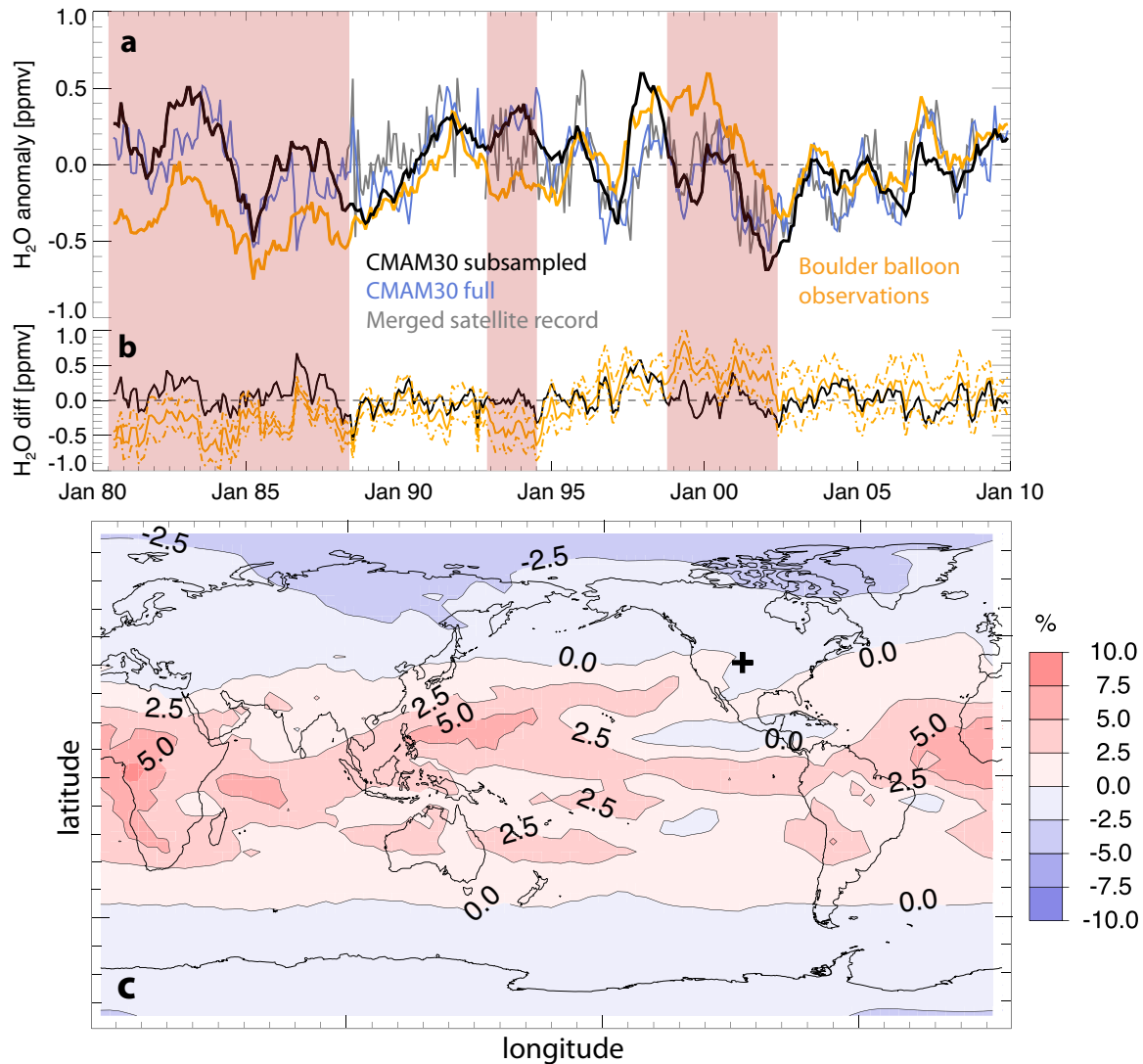
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Figure 1: Approach to merging a climate data record. Timeseries of monthly zonal mean water vapour at 100 hPa averaged over 20S-20N for 1988-2010: **(a)** Absolute mixing ratios from different instruments (colours) and CMAM30 (grey), **(b)** differences and **(c)** bias-corrected differences between observations and CMAM30, and **(d)** bias-corrected absolute mixing ratios from observations. Grey solid and dashed horizontal lines in (d) indicate mean and 1-sigma standard deviation of the observational record averaged over the whole time period. The red box encompasses months excluded from the relative-bias determination due to identified problems in ERA-Interim (see text). See Supplementary Material for SCIAMACHY bias explanation.



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Figure 2: Consistency between tropical tropopause temperature and lower-stratospheric water vapour. Scaled anomalies (unitless) of tropical temperature at 100 hPa averaged over 15S-15N and of water vapour at **(a)** 80 hPa averaged over 20S-20N and **(d)** 100 hPa averaged over 40N-50N. Tropical temperature is lagged by 2 (3) months in the tropics (extratropics). **(b, c, and d)** Differences between scaled temperature and water-vapour anomalies (unitless). Red bar highlights a time period where the scaled anomalies in the model and the observations show a substantial disagreement, blue bars where the temperature-water vapour relationship is strongly perturbed in both model and observations.



389

390

391 **Figure 3: Comparison of stratospheric water vapour from Boulder balloon and**392 **merged satellite datasets. (a)** Deseasonalized water-vapour anomalies at 100 hPa

393 derived from Boulder balloon observations (orange), the zonal mean (40N-50N)

394 model (blue) and merged satellite data (grey), and the model subsampled at Boulder

395 (black). **(b)** Differences between full model anomalies and balloon (orange) or sub-

396 sampled model anomalies (black). Red shadings highlight periods where sub-

397 sampled model data systematically lie outside the 1-sigma uncertainty of the

398 balloon observations (dashed orange lines). **(c)** Longitude-latitude percentage

399 changes of water vapour at 100 hPa for 1980-2010 from the model. The cross

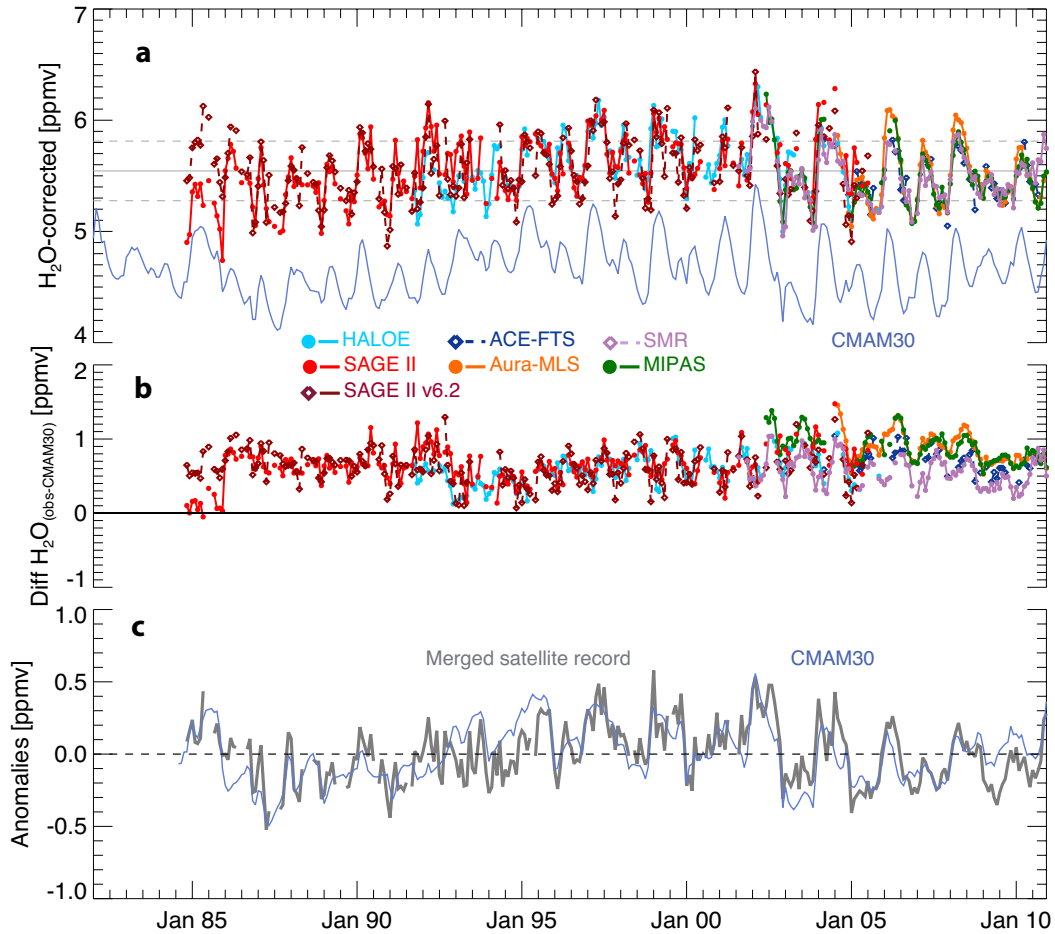
400 indicates the location of Boulder (40N/105W).

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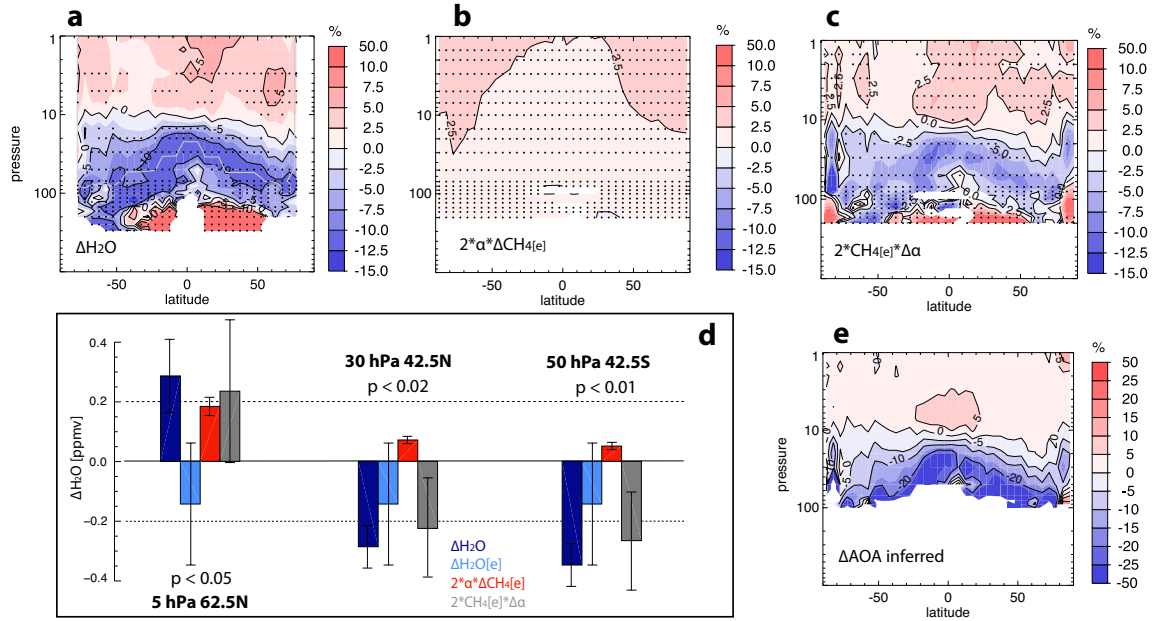
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Figure 4: Extension of the water-vapour timeseries back to the mid 1980s. (a) Timeseries of zonal mean water vapour at 10 hPa and 40N for model (grey) and the different instruments (bias-corrected and colour-coded). **(b)** Relative biases between each instrument's original monthly zonal mean timeseries and CMAM30. **(c)** Deseasonalized anomalies of the merged satellite water-vapour record (grey) and the model.



414
 415 **Figure 5: Long-term changes in stratospheric water vapour and its drivers. (a)**
 416 Percentage changes up to 2010 derived from the merged satellite record since
 417 1986/1988 above/below grey line. Dots indicate 95%-significance level.
 418 Contribution to (a) from (b) tropospheric methane increases and (c) inferred
 419 changes in the stratospheric circulation. (d) Absolute water-vapour changes at
 420 three locations and contributions from their drivers including uncertainties (as
 421 discussed in detail in *Supplementary Material*). P-values are given for the difference
 422 between observed changes and the sum of the two entry-value contributions. (e)
 423 Fractional-release factor changes translated into age-of-air (AOA) changes
 424 (significance not estimated).

425
 426
 427 **Table 1: (a) Water vapour and (b) temperature changes derived for different**
 428 **time periods, latitude bands, and altitudes from observations and from**
 429 **CMAM30.** Changes are calculated for the timeseries shown in *Figures 2, S1, 3, and 5*
 430 and given as total change in ppmv over the entire period. Most trends are not
 431 statistically significant due to large variability in stratospheric water vapour when
 432 compared to data record length. Decadal fluctuations strongly affect derived trend
 433 values depending on the time period chosen^{10,31}. Trend and significance calculation
 434 is explained in the Methods section. *CMAM30ss* denotes sub-sampled model fields.

435

H ₂ O	Period	Lat Band	Height [hPa]	Change [ppmv]	2-sigma Uncertainty [ppmv]	t-value	Effective Sample #	Significance Level
Figures 2 and S1								
Satellite	1988-2010	20S-20N	80	-0.14	0.20	0.67	26	70%
Satellite	1988-2010	20S-20N	100	0.01	0.14	0.06	53	Not sig

Figure 3								
Satellite	1988-2010	37.5-42.5N	100	-0.05	0.17	0.27	32	Not sig
Boulder	1988-2010	37.5-42.5N	100	0.39	0.18	2.17	129	95%
CMAM30ss	1988-2010	37.5-42.5N	100	-0.15	0.22	0.66	84	Not sig
CMAM30	1988-2010	37.5-42.5N	100	-0.07	0.22	0.32	16	Not sig
Figure 3 (continued)								
Boulder	1980-2010	37.5-42.5N	100	0.60	0.15	3.9	160	95%
CMAM30ss	1980-2010	37.5-42.5N	100	-0.27	0.18	1.49	110	90%
CMAM30	1980-2010	37.5-42.5N	100	-0.12	0.19	0.64	22	Not sig
Figure 5e								
Satellite	1986-2010	62.5N	5	0.28	0.12	2.4	26	95%
Satellite	1986-2010	42.5N	30	-0.28	0.07	4.1	65	95%
Satellite	1986-2010	42.5S	50	-0.34	0.07	5.2	72	95%
Temp	Period	Lat Band	Height [hPa]	Change [K]	2-sigma Uncertainty [K]	t-value	Effective Sample #	Significance Level
Figure 2								
Model	1988-2010	15S-15N	100	-0.04	0.32	0.13	249	Not sig

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439 **Methods**

440

441 ***Nudged Chemistry-Climate Model simulations***. The CMAM30 dataset is produced
442 using the Canadian Middle Atmosphere Model⁴⁰ driven by the latest European
443 Centre for Medium-Range Weather Forecasts (ECMWF) ERA-Interim reanalysis²¹
444 over the past 30 years (1980-2010). For details on the nudging see *Supplementary*
445 *Material*. The model was run on 71 vertical levels from the surface to around 95 km,
446 with a vertical resolution of approximately 1 km around the tropopause, and a
447 horizontal resolution of T47, or approximately 4 degrees. The stratospheric source
448 gas of water vapour, methane, is prescribed as a time-varying, global average
449 surface concentration based on observations and is subject to model transport and
450 chemistry. Water vapour is likewise a fully prognostic field in the model, chemically
451 produced by methane oxidation and removed through parameterized large-scale
452 and deep convective precipitation processes. Water vapour in excess of the local
453 saturation mixing ratio is removed, following the rationale that the stratospheric
454 water-vapour entry value is largely determined by the Lagrangian cold point as air
455 passes through the tropical tropopause¹⁹, but neglecting super-saturation⁴¹. The
456 free-running CMAM has been evaluated for its performance in the upper
457 troposphere and lower stratosphere both in the tropics¹² and extratropics⁴² and
458 found to be one of the best-performing models. CMAM30 data can be downloaded
459 from [http://www.cccma.ec.gc.ca/data/cmam/output/CMAM/CMAM30-](http://www.cccma.ec.gc.ca/data/cmam/output/CMAM/CMAM30-SD/index.shtml)
460 [SD/index.shtml](http://www.cccma.ec.gc.ca/data/cmam/output/CMAM/CMAM30-SD/index.shtml)

461

462 ***SPARC Data Initiative timeseries***. The SPARC Data Initiative water-vapour
463 timeseries have been compiled using profile data that were carefully screened
464 before binning, and a hybrid log-linear interpolation in the vertical has been
465 performed. The timeseries feature zonal monthly mean cross sections with a
466 horizontal resolution of 5° on 28 pressure levels between 300 and 0.1 hPa (around
467 64 km altitude). We here use the timeseries from seven instruments, which provide
468 near-global coverage (SAGE II, HALOE, Odin/SMR, SCIAMACHY, ACE-FTS, Aura-MLS,
469 and MIPAS) and have been quality-assessed within the SPARC Data Initiative²³.
470 Sampling issues are discussed in Ref 43. The climatologies were based on the
471 following data versions (specific references are provided in Ref 23): HALOE v19,
472 SMR v2.0 (in the lower stratosphere) and SMR v2.1 (in the middle and upper
473 stratosphere), SCIAMACHY v3.0, ACE-FTS v2.2, Aura-MLS v3.3, and MIPAS
474 v3o_H2O_13 (for 2002-2004 data) and v5r_H2O_220 (for 2005-2010 data, where
475 the operation mode was switched from high-spectral to low-spectral resolution).
476 SAGE II v6.2 submitted to the SPARC Data Initiative is only shown in *Figure 4*, but
477 otherwise is superseded by climatologies based on the improved SAGE II v7.0
478 data⁴³. For methane, ACE-FTS data⁴⁵ were used. The SPARC Data Initiative
479 climatologies can be downloaded from [http://www.sparc-climate.org/data-](http://www.sparc-climate.org/data-center/data-access/sparc-data-initiative/)
480 [center/data-access/sparc-data-initiative/](http://www.sparc-climate.org/data-center/data-access/sparc-data-initiative/)

481

482 ***Boulder balloon observations***. Water vapour vertical profile measurements over
483 Boulder by balloon-borne NOAA frost point hygrometers (FPHs) started in April
484 1980 and continue today. Most soundings were conducted monthly, however the

485 record contains several multi-month data gaps, especially above 22 km. A
486 comparison of FPH and Aura-MLS measurements over Boulder and Lauder, New
487 Zealand, shows no significant temporal drifts between the two instruments from
488 100 to 26 hPa during 2004-2012 (Ref. 46). See also discussion of measurement
489 uncertainty in the *Supplementary Material*.

490

491 **Anomalies.** Anomalies are calculated with respect to the full time period depicted in
492 the different figures, by subtracting the seasonal cycle derived from each individual
493 instrument or the model from the overall time series.

494

495 **Trends and significance tests.** Unless indicated otherwise, uncertainty estimates
496 are given as two sigma throughout the manuscript. We use a least-square linear
497 regression in order to derive the trends from deseasonalized water-vapour anomaly
498 timeseries at the different altitudes and latitudes. The significance of the trend is
499 derived taking into account the effect of potential autocorrelation within the
500 timeseries on the number of independent data points (reducing the effective sample
501 size). This effective sample size is then used to recalculate the uncertainty of the
502 derived trends and determine the tabulated one-sided student's t-test value, used to
503 define the significance level of the trends. A more detailed discussion of the method
504 can be found in Ref 47.

505

506 **Total-water diagnostic.** Apart from polar dehydration and other non-conservative
507 processes, stratospheric "total water" $\text{H}_2\text{O} + 2 \cdot \text{CH}_4$ is approximately conserved^{33,34,16}
508 hence water vapour and methane at a given location can be written as

509

$$510 \text{H}_2\text{O} = \text{H}_2\text{O}_{[e]} + 2\alpha\text{CH}_{4[e]}, \quad \text{CH}_4 = (1-\alpha) \text{CH}_{4[e]} \quad (1a,b)$$

511

512 where the subscript '[e]' refers to the entry value at the tropical tropopause (lagged
513 by the mean age-of-air), and α represents a fractional-release factor which depends
514 on the circulation. Under this assumption, sufficiently small water-vapour changes
515 can be attributed to changes in water-vapour entry value, methane entry value, and
516 circulation according to

517

$$518 \Delta\text{H}_2\text{O} = \Delta\text{H}_2\text{O}_{[e]} + 2\alpha\Delta\text{CH}_{4[e]} + 2\text{CH}_{4[e]}\Delta\alpha. \quad (2)$$

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520 The calculation of α and $\Delta\text{CH}_{4[e]}$ and their uncertainties is described in the
521 *Supplementary Material*. The last term in (2) is calculated as a residual of the other
522 terms that can be derived from observations with its uncertainty being
523 overwhelmingly dominated by that of $\Delta\text{H}_2\text{O}_{[e]}$. Hence the other uncertainties
524 (including possible non-conservation of total water) are not critical.

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