

Interactive comment on “Water Vapour and Methane Coupling in the Stratosphere observed with SCIAMACHY Solar Occultation Measurements” by Stefan Noël et al.

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We thank the reviewer for the detailed comments which will help us to improve the paper. In the following, the original reviewer’s comments are given in *italics*, our answer in normal font and the proposed updated text for the new version of the manuscript in **bold** font.

Note: It seems that line numbers in the comments refer to the manuscript before technical corrections, not the published discussion version.

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General Comments

- *The paper is generally well written, but some of the grammar and phrasing could be significantly improved by allowing a native English speaker to edit it.*

We will try to improve the English in the revised version and let our English co-author check it again. Therefore, the updated text might change slightly in the final revised version.

- *The oxidation of hydrogen (H₂) in the stratosphere is also a source of water vapor. How is it that the equation for total hydrogen (“potential water”) does not include a term for H₂ oxidation?*

We use the definition of Nassar et al. (2005) for potential water which does not include H₂ (assuming that variations of H₂ are small). This is mentioned in the introduction.

- *The paper would benefit from an early paragraph dedicated to describing the influences of the QBO on stratospheric entry mixing ratios of H₂O and on the conversion of CH₄ to H₂O during transport from the tropics to higher latitudes. Currently there is a lot of attribution of inter-annual variations in H₂O and CH₄ to the QBO, but the actual QBO mechanisms that drive these variations are not really mentioned until late in the paper. Similarly, an earlier introduction of concepts like the lower and upper branches of the Brewer-Dobson circulation, along with a description of mean age and its utility in discerning stratospheric transport pathways, would be very helpful when discussing the observed couplings between CH₄ and H₂O. Currently, these concepts are discussed too late in the paper. The reasons why total hydrogen is a conserved quantity above the lowermost stratosphere may escape some readers. A simple explanation should be given, perhaps illustrating how mixing between air masses during transport does not change total hydrogen.*

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Will add a corresponding paragraph in the introduction, which will be largely re-written in the revised version.

- *I don't see the need for Figure 11 as I am not quite sure what it explains. There is no caption to describe what is meant by the different shadings of green and purple (and white) arrows. I don't see anything in this Figure that isn't already described in detail in the text.*

Indeed, this figure does not contain additional information about the results, but we think it is helpful to visualise the different transport pathways and related processes. We therefore prefer to keep the figure in the manuscript, but move it to the introduction. Different shadings are mainly for artistic purpose and should illustrate dynamics.

- *All trend values in the text should be presented with their uncertainties (95% confidence intervals) so the reader can gauge their significance. At many altitudes (if not all), the 9-year trends of CH₄, H₂O and total hydrogen are not statistically significant. Labeling trends as “negative”, “positive”, or “near-zero” is not justified if they are not statistically different from zero.*

We will add trend uncertainties (2σ values) and also adapt the text accordingly (see answers to specific comments below).

Specific Comments

- *Page 1, Line 4: “theses” should be “these”*
Will be corrected.
- *P1, L6-8: Are these trends “significant” as stated? Please include their uncertainties to show that they are statistically different from zero.*

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The mentioned trends are significant. Uncertainties (about 0.008 ppm/year, see also Fig. 10) will be added.

- *P1, L10: “are strongly correlated” should be “are strongly anti-correlated”*

Will be corrected.

- *Pages 1 and 8 have the QBO erroneously defined as the “Quasi-Biannual Oscillation” instead of “Quasi-Biennial Oscillation”. You also use the term “bi-annual structure” on pages 8 and 11, where I assume you mean “biennial”, since “bi-annual” means every 6 months.*

The referee is absolutely right – it should be “biennial” in all cases, sorry for this mistake. We will correct this.

- *P1, L11: Why does it only “seem” that most of the water vapor is produced by methane? What else might produce water vapor above 20 km?*

There are in fact other sources of stratospheric water vapour under discussion, e.g. from aviation or volcanoes. However, this is mainly relevant for the lowest parts of the stratosphere, therefore we will change this sentence to:

Above about 20 km most of the water vapour is attributed to the oxidation of methane.

- *P1, L13-15: Why should there be a “balance between water vapor and methane” at lower altitudes, unless of course the air masses came from higher in the stratosphere where there is a photochemical balance between CH₄ and H₂O? But here you mention only “the lower branch of the Brewer-Dobson circulation” where this photochemical balance does not exist. I don’t understand the intention of this sentence and, to me, it is confusing.*

We agree that this may be confusing and will reformulate the sentence:

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The SCIAMACHY data confirm, that at lower altitudes the amount of water vapour and methane are transported from the tropics to higher latitudes via the shallow branch of the Brewer-Dobson circulation.

- *P1, L17-18: It is too strong to say that these three greenhouse gases “determine the climate on our planet” when there are many, many contributors to Earth’s climate.*

Agreed. We will reformulate this to:

Water vapour (H₂O), methane (CH₄) and carbon dioxide (CO₂) are all greenhouse gases.

- *P1, L22: If “methane is mainly produced in the troposphere”, where else is it produced?*

There are indeed no known stratospheric sources of methane. We will therefore remove “mainly”.

- *P1, L23: For decades, there have been attempts to produce spatially-resolved estimates of CH₄ sources using globally-distributed surface measurements and inverse models. This is not something novel. How are satellite measurements used to identify methane sinks?*

Several satellite instruments (including SCIAMACHY, but also GOSAT and soon TROPOMI on Sentinel 5p) provide CH₄ data, usually total columns determined from nadir measurements. These can be used in combination with inversion models to derive sources and sinks. The referee might have a look at the GHG-CCI web site (<http://www.esa-ghg-cci.org>) for more information about available data sets.

- *P2, L1: What is the “long” lifetime of “tropospheric methane”? Be more quantitative.*

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The lifetime of tropospheric methane is about 10 years, we will mention that.

- *P2, L5: There were papers published long before 2001 that describe the “cold trap”. In fact, there was some pioneering work performed back in the 1940s by Brewer and Dobson.*

It is true that the “cold trap” has been discussed before 2001. We only wanted to give some example references here. We will add as additional (early) example the work by Brewer(1949), see reference in Holton and Gettelman (2001).

- *P2, L8: What is the connection between “tropical upwelling” and the “freeze-drying process”?*

Tropical upwelling transports air masses from the troposphere into the stratosphere. As mentioned before, the tropopause acts as a cold trap such that water vapour partly freezes out before reaching the stratosphere, which is therefore dry compared to the troposphere. We will explain this in the revised version.

- *P2, L10: Why only “in the middle stratosphere and above” is water vapor “produced from (the) oxidation of stratospheric methane”? Both le Texier et al. (1988) and Rohs et al. (JGR, 2006) clearly show that some methane is oxidized in the lower stratosphere.*

In the lower stratosphere oxidation of methane is not the only source of water vapour, there is e.g. also a tropospheric source (as we discuss in the present manuscript). However, as this sentence may be misleading, we will replace “in the middle stratosphere and above” by “in the stratosphere”.

- *P2, L17: The concept of potential water, historically referred to as “total hydrogen”, being conserved in stratospheric air masses as they mix and photochemically age, has been known for a long time. It pre-dates Rinsland et al. (GRL, 1996), so citing a 2005 paper here ignores the pioneering work on this topic that was performed well before the 21st century.*

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The concept of potential water is indeed older than the mentioned publication from Nassar et al. (2005). However, Nassar et al. (2005) define the term “potential water” in contrast to “total hydrogen” (which includes H₂) as we use it in our manuscript, therefore we cite this paper here, but we will also add the Rinsland reference.

- *P2, L19-21: It is not “the combination”, but rather “simultaneous measurements of” H₂O and CH₄ profiles, that are useful in understanding the connection between the two gases. Why is it best that they be measured by the same instrument? Does this improve the accuracy of H₂O and CH₄ retrievals, and therefore total hydrogen values?*

If measurements from the same instrument (and similar retrievals) are used, possible systematic effects caused by the instrument or the retrieval method may cancel. This should improve the accuracy of the resulting potential water / total hydrogen.

We will reformulate this:

Ideally, both water vapour and methane should be retrieved from measurements by the same instrument. In this case, the collocation of the two data sets is very close. Furthermore, possible systematic errors caused e.g. by instrument calibration or by the retrieval method may to some extent cancel.

- *P5, L9-11: A “criterium” is a bicycle race. Instead use “criterion” (singular form of criteria). I’m not sure what “a maximum time distance of 9 hours” means. And does “the closest match” refer to time or distance?*

We will replace “criterium” by “criterion”. “a maximum time distance of 9 hours” refers to the difference between the measurement times of the two instruments; “the closest match” refers to spatial distance. For clarification, we will reformulate the corresponding sentence:

For MLS we use a maximum time distance of 9 hours between MLS and SCIAMACHY measurements and always take the spatially closest match.

- *P5, L15-17: What version of MLS retrievals are you using? Hopefully the latest and greatest, v4.2. The phrases “slightly higher” and “typically smaller” convey very little information. Please be more quantitative.*

We indeed use MLS V4.2 and will mention this in the text and the related figure caption. We will also give quantitative numbers in the related sentence:

The SCIAMACHY water vapour VMRs are usually about 2–3% higher than those of ACE-FTS, but (except for the lowest altitudes) typically 2–3% smaller than MLS VMRs.

- *P6, Figure 3: I would be careful when using the term “absolute differences” because “absolute” may infer absolute values. Given the x-axis units (ppmv or %) I think it is safe to remove “absolute” and “relative” from the Figure headings.*

“absolute” might indeed be misleading as we show positive and negative values and can be removed. We will modify Figs. 3 & 4 and their captions accordingly. We see however no problem with “relative” and would prefer to keep this in order to better distinguish panels a) and b).

- *P6, Figure 3d: Why does this vertical profile of correlation coefficients for SCIA vs ACE have such an altitude-dependent shape? The scatter in SCIA-ACE differences (ppmv and %) does increase somewhat near the lower and upper altitude boundaries, but is this enough to decrease the correlation coefficients near 17 and 45 km by more than a factor of two from those in the 25-40 km range? Do the correlation coefficients decline because of diminishing data populations as the altitude boundaries are approached? Figure 4d has a similar shape, but the r values don't fall so severely as the boundaries are approached. What makes these panel (d) curves similar in shape but so different in r values near the altitude boundaries?*

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The possible reason for the decreasing r in Fig. 3 at lower altitudes is that the variability of the ACE-FTS data is higher than for SCIAMACHY. This can be seen from the standard deviations shown in panel c). High correlation is achieved when variability (standard deviation) is similar for both data sets, i.e. in this case both instruments see the same atmospheric changes. MLS standard deviation is at lower altitudes closer to that of SCIAMACHY, therefore the correlation is higher.

We suggest to add the following text to explain this:

The correlation between SCIAMACHY and both ACE-FTS and MLS data is generally high (reaching about 0.85 at 30km), but is poorer at lower and higher altitudes. The reduction at higher altitudes may be a consequence of the larger relative errors of the SCIAMACHY data, but as yet there is no clear explanation. At lower altitudes, differences in the variability of the data play a role, as can be inferred from the standard deviations shown in panels c) of Fig. 3 and 4. High correlation is achieved when variability and variance are similar for both data sets, i.e. in this case both instruments see the same atmospheric changes.

- *P8, L3-5: What is meant by “bi-annual structure” in Figure 6? I don’t see any cycles in the H₂O or CH₄ anomalies that clearly repeat with a 6-month (biannual) or 2-year (biennial) period. I do see lots of inter-annual variability. Is that what you want to say? Also, why does one expect inter-annual variability in CH₄ and H₂O because of the QBO? What are the mechanisms that drive changes in both?*

We mean “biennial structure”, i.e. a variation with a 2-year period. This is seen especially at altitudes around 25–30 km where red and blue patterns repeat about every two years. This is seen more clearly in the following figures; related mechanisms are discussed in the “Discussions” section later.

- *P8, L6-8: “show an inverted behavior”. Do you instead mean “opposite behavior” since “opposite” implies negative vs positive? The water vapor anomalies are*

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“about twice as high negative” is awkwardly worded. How about “The methane anomalies correspond to water vapor anomalies that are opposite in sign and twice the magnitude.” Also, the statement “that most of the water vapor is produced from methane” is not correct since mixing ratios of H₂O are ~4 ppmv at stratospheric entry and ~7 ppmv at 45 km.

Yes, we mean “opposite” and will change the text. “most of the water vapor is produced from methane” actually refers to anomalies, i.e. changes in water vapour and methane, we will clarify that.

New text:

The methane anomalies correspond to water vapour anomalies that are opposite in sign and twice the magnitude. This complies with the assumption, that most of the changes in water vapour are produced from methane via the net reaction (R2).

- *P9, L2-3: Don't the water vapor anomalies at 17 km also show year-to-year differences in the amount of water passing through the tropical cold trap, i.e., variability not related to the QBO? By how many months is the QBO signal at 17 km “shifted in phase” from that at 25 km? Is the reason for this phase shift that the QBO propagates downward?*

Water vapour entering the stratosphere in the tropics varies also due to a combination of QBO and BDC effects. However, our measurements indicate that QBO effects dominate in this case. We try to explain this in the “Discussions” section later in the manuscript. According to our explanation, the air at 17 km is several years younger than the air above about 25 km (because of the different pathways of the Brewer-Dobson-Circulation). The phase shift between 17 and 25 km is therefore not only a few months but probably more than one 2-year period, and it is not possible to determine the exact value from our data. Above about 25 km there are indeed some indications for downward transport, as can be seen in the

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slanted structures of the anomalies shown in Fig. 6.

We will add the following text for clarification:

Note that the age of air at these altitudes may be up to about 8 years according to e.g. Haenel et al. (2015). Consequently, the actual phase shift is expected to be larger than one 2-year period of the QBO. It therefore cannot be determined well from our 9-year time series.

- *P9, L4: “downward peak” is contradictory. How about “dip”? I presume here you are still discussing the 17-km data?*

We will reformulate the sentence to clarify this:

The dip in the water vapour anomalies at 17 km in the middle of 2009 is related to the eruption of the Sarychev volcano...

- *P9, L10: I would call the Singapore zonal wind average a QBO “index” rather than a “proxy”.*

OK, will be changed.

- *Figure 8 caption: Not only is the y-axis for CH₄ inverted, it is also scaled differently than the y-axis for H₂O.*

The caption will be changed accordingly:

Note that the vertical axis of the methane data is inverted and scaled differently than for water vapour.

- *P10, L6-8: What is the average transport time from the tropics to the northern 50-70° latitude at 30 km? You could determine this by independently regressing the H₂O and CH₄ anomalies against U10 and progressively delaying the U10 index one month at a time, finding the delay that produces the highest correlation coefficients. On Line 8 you say “positive anomaly in the wind data”, but Figure*

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8 doesn't show wind anomalies. It is strange that the positive anomalies in H₂O and CH₄ at the beginning of 2010 were not preceded by positive zonal mean winds.

Age of air at these altitudes is about 8 years (see manuscript and above). Therefore the delay between tropospheric winds and stratospheric H₂O or CH₄ is more than one QBO period. We think the time series is too short and does not contain enough distinct features to determine these large delay times.

We will change “positive anomaly” to “positive values” as these are indeed no anomalies:

The positive values in the wind data around 2010/2011 are hardly detected in the methane and water vapour data.

The behaviour after 2010 is indeed strange and needs further investigations, as we mention in the text.

- *P11, L1: Water vapor is also produced by the oxidation of hydrogen (H₂) in the stratosphere. How does this factor into $H_2O + 2*CH_4 = \text{constant}$?*

Indeed, H₂ needs to be considered in the sum as only total hydrogen is conserved. However, as mentioned in the introduction, for potential water we assume that H₂ variations can be neglected.

- *P11, L4-5: Why is the QBO signal visible only below 20 km in Figure 9? What mechanism alters $H_2O + 2*CH_4$ below 20 km but not above this altitude? Only Figure 7d shows greater variations in H₂O anomalies than in CH₄ anomalies.*

We discuss this in the “Discussions” section. The basic idea is that H₂O at higher altitudes is produced from CH₄ such that the combination does not show a QBO signal. At lower altitudes, H₂O shows a QBO signal caused by variations due to QBO effects on tropopause temperature. CH₄ transport into the stratosphere is

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not affected by tropopause temperature changes and therefore does not show a QBO signal.

- *P11, L11-13: What could possibly drive changes in $H_2O + 2*CH_4$ with a periodicity of 5-6 years? I don't think this statement is supported by Figure 9 that spans only 9 years.*

There is no explanation for this 5–6 years periodicity yet. Possible reasons are variations in the Brewer-Dobson circulation or changes in water vapour trends; we will mention this in the discussion. We also agree that it is difficult to tell if this periodicity is real from our data.

To clarify this we will reformulate this sentence to:

This implies a periodicity of about 5 to 6 years, but due to the limited length of the time series, this can only be confirmed in the future.

- *P11, L15: I don't see the that scatter (std dev) in SCIAMACHY retrievals increases significantly above 40 km (Figures 3c and 4c), so what do you mean here by "large uncertainties of the ONPD data at higher altitudes"?*

This refers to the (mean) error on the data which increases with altitude, see Figs. 3 & 4 panels a) and b).

- *P11, L17-18: Please expand your description of the linear trend fitting here, at least in a general way. At what altitudes did you determine trends? Did you perform any vertical averaging (other than averaging kernels) of the profiles before determining trends? There is not enough information presented here to simply reference an earlier paper.*

We will modify the text to describe the fitting procedure further:

To derive these changes, a linear regression has been fitted to the water vapour anomalies at each altitude similar to that used in the earlier methane

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study, see Noël et al. (2016). For this, we take the anomaly times series at a selected altitude (see e.g. Fig. 7) and fit a straight line to it. The slope of this line is the estimated trend for this altitude, the error of the trend is the error of the slope given by the fit. This procedure is undertaken at all altitudes from 17 to 45 km, in 1 km steps. The resulting trend profiles are displayed in Fig. 10.

- *P11, L20: Here and throughout, all trends need to be presented with their 95% confidence intervals. Otherwise, the reader has no idea if the trends are statistically significant or not unless they check Figure 10. Many of the trends between 25 and 40 km are NOT negative, they are indistinguishable from zero. Only the H₂O trends between 31 and 37 km can be labeled as negative.*

The derived values of the H₂O trends between about 25 and 40 km are negative, but it is true that some of these trends are not significant. We explain in the text which regions are significant and which are not. For further clarification, we will add the uncertainties to the trends mentioned explicitly in the text.

- *Figure 10: Please scale the x-axis for CH₄ accordingly for $d[\text{H}_2\text{O}]/dt + 2 \cdot (d[\text{CH}_4]/dt) = 0$. Wouldn't one expect a positive trend in CH₄ accompanying the negative trend in H₂O between 31 and 37 km?*

We will scale the x-axis of the CH₄ plot in Fig. 10 by a factor of 2. One would indeed expect a positive trend for CH₄ between 31 and 37 km, but the resulting errors on the trends are high, so the CH₄ trends and also the combined PW trends are not significant.

- *P12, L7-8: Please remove one of the repeated "an estimate"*
OK.
- *P12, L12: "not disproved" is a very weak way to say this. How about "Given that the trends in potential water between 21 and 45 km lack statistical significance,*

there is no evidence that water vapor is produced in the stratosphere by any mechanism other than methane oxidation.”

OK, will be changed.

- *P12, L13: “where the trend itself is close to zero” is not supported by the 95% confidence intervals of -0.015 to +0.014 ppmv/year in the 25-30 km altitude range.*

Agreed. Although the value is close to zero the trend is not significant, we will remove this sentence.

- *P12, L19-20: Why is this? You haven’t explained why the QBO might influence H₂O but not CH₄ in the lower stratosphere. You also haven’t explained why there should be a lag between QBO water vapor signals in the upper and lower stratosphere. Is it a difference in the mean ages of the air masses? It would be a good idea to introduce the concept of mean age early in this paper if you are going to discuss differences in the “phasing” of QBO-induced water vapor signals at different altitudes.*

An explanation for the observed features in this list is given in the subsequent paragraphs in the manuscript. We will reformulate this section and add some additional information about age of air in the introduction (see answer to general comments).

- *P13, L5-9: I think this explanation should appear earlier in the paper. This is not a conclusion of the paper, it is information pertinent to the understanding of why QBO “signals” in H₂O at different altitudes are present at different times.*

We will add some information about the different branches in the introduction:

There are in principle two pathways for this transport (see e.g. Butchart, 2014, and references therein): At lower altitudes, air masses are transported via the shallow (or lower) branch of the Brewer-Dobson circulation.

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At higher altitudes the water vapour is transported by the deep (or upper) branch of the Brewer-Dobson circulation.

- *P13, L11: “water vapor is mainly produced from methane oxidation”. What else produces water vapor at these altitudes? Also, I think you need to have a definitive statement earlier in the paper that the oxidation of methane to water occurs predominantly in the tropical stratosphere and the fraction of methane converted to water increases with altitude.*

We will remove “mainly” and add some more information in the introduction.

- *P13, L15-17: Don't forget the main driver of variability in stratospheric H₂O entry mixing ratios is the seasonal cycle of tropical tropopause temperatures. Also, ENSO can significantly influence water vapor input to the tropical lower stratosphere by affecting tropical tropopause temperatures and through convective activity. A lack of strong seasonal, QBO and ENSO influences on UTLs methane DOES explain the lack of CH₄ variability at 17 km.*

Since we are looking at anomalies here, seasonal cycle effects should be removed. During the period of SCIAMACHY measurements there were no strong ENSO events, so this impact should be limited. Therefore we think that the missing QBO influence is a valid (and in this specific case sufficient) explanation for the lack of CH₄ variability at 17 km.

- *P14, L5-7: As per my previous comment about introducing the concept of mean age, here at the end of the paper is just such an introduction. I think the paper would benefit from this appearing much earlier.*

We will add some sentences on age of air in the introduction (see answer to general comments).

- *P14, L10: This sentence makes it sound like CH₄ was emitted at 17 km. And is mean age really the elapsed time from emission, including transport time from*

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extra-tropical sources to the tropics?

The formulation is indeed misleading. We will change this to:

about 2–3 years between injection into the stratosphere at the tropics and measurement at 17 km at higher latitudes

- *P14, L17: The concept of “QBO signal has to be carried by methane” is an awkward way of explaining QBO influences on the oxidation of CH₄ to H₂O. If the QBO can alter the strength of the Brewer-Dobson circulation then it can also change the amount of CH₄ oxidized to H₂O during poleward transport. Transport times depend on the strength of the B-D circulation because this can also alter the path (i.e., stronger = higher path) and therefore the amount of CH₄ oxidized to H₂O. I think a paragraph early in the paper should be dedicated to HOW the QBO affects stratospheric transport and therefore the amount of CH₄ converted to H₂O during transport from the tropical lower stratosphere to the higher latitudes of your data set.*

We will add a corresponding part in the introduction, see answer to general comments.

- *P14, L29-30: Please include trend uncertainties with the trends.*
Will be done.
- *P15, L1: “At altitudes above about 20 km, variations in water vapor . . .”*
Will be added.
- *P15, L6: Why is potential water not constant over time? Were there changes in the stratospheric entry mixing ratios of H₂O? Of CH₄? Of both?*

Actually, we do not know the reasons why potential water varies on a timescale of 5–6 years, but we will mention possible reasons (low-frequency variations in the Brewer-Dobson circulation or in water vapour trends) in the discussion. Our

data set does not extend to the tropics, therefore we cannot infer changes of the entry mixing ratios.

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