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Atmospheric Chemistry and Physics Discussions

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

Anonymous Referee #1

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This paper examines SCIAMACHY measurements of stratospheric water vapor and methane in the 50°N-70°N zonal band during 2002-2012. The focus is how stratospheric methane and water vapor abundances above about 20 km altitude are linked by the reactions that oxidize methane to water vapor. In this atmospheric region, the quantitative conversion of methane to water vapor makes total hydrogen (H2O+2*CH4) a conserved quantity as air masses mix and photochemically age. Inter-annual changes in H2O and CH4 anomalies at various altitudes are attributed to the quasi-biennial oscillation (QBO). The data are also used to determine 9-year linear trends in H2O, CH4 and total hydrogen at stratospheric altitudes between 17 and 45 km. Most of the trends are not statistically significant. QBO effects on the relationship between H2O and CH4

are discussed in the context of the upper and lower branches of the Brewer-Dobson circulation, for which the effects are different.

========== 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.

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?

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.

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.

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.

========== Specific Comments ==========

Page 1, Line 4: “theses” should be “these”

P1, L6-8: Are these trends “significant” as stated? Please include their uncertainties to show that they are statistically different from zero.

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

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.

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?

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.

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.

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

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?

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

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.

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

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.

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.

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?

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?

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.

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.

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?

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?

P8, L6-8: “show an inverted behavior”. Do you instead mean “opposite behavior” since “opposite” implies negative vs positive? The water vapor anomalies are “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 $\sim$4 ppmv at stratospheric entry and $\sim$7 ppmv at 45 km.

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?

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

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

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

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 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.

P11, L1: Water vapor is also produced by the oxidation of hydrogen (H2) in the stratosphere. How does this factor into H2O + 2*CH4 = constant?

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

P11, L11-13: What could possibly drive changes in H2O + 2*CH4 with a periodicity of 5-6 years? I don’t think this statement is supported by Figure 9 that spans only 9 years.

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”?

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.

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.

Figure 10: Please scale the x-axis for CH4 accordingly for d[H2O]/dt + 2*(d[CH4]/dt) = 0. Wouldn't one expect a positive trend in CH4 accompanying the negative trend in H2O between 31 and 37 km?

P12, L7-8: Please remove one of the repeated "an estimate"

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."

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.

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.

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.

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.

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.

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.

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 extra-tropical sources to the tropics?

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.

P14, L29-30: Please include trend uncertainties with the trends.

P15, L1: “At altitudes above about 20 km, variations in water vapor . . .”

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?

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