

Interactive comment on “Molecular hydrogen (H₂) combustion emissions and their isotope (D/H) signatures from domestic heaters, diesel vehicle engines, waste incinerator plants, and biomass burning” by M. K. Vollmer et al.

Anonymous Referee #1

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Title: Molecular hydrogen (H₂) combustion emissions and their (D/H) signatures from domestic heaters, diesel vehicle engine, waste incinerator plants, and biomass burning
Authors: Vollmer et al.

The submitted manuscript presents measurements of hydrogen (H₂), its isotope signature (dD), carbon monoxide (CO), carbon dioxide (CO₂) and methane (CH₄) sampled from a wide range of combustion sources.

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This manuscript reports detailed analysis of samples from all major emission sources of anthropogenic H₂: domestic oil, gas and wood heaters, diesel vehicle engines and waste incinerators. Using the H₂/CO ratios of these sources detailed global emission estimates of H₂ are reported. A new method of calculation of H₂ emissions is also used in this manuscript where the H₂/CH₄ ratio from biofuel and biomass burning is used with CH₄ inventories to estimate global H₂ emissions.

My general comment on this paper is that it deserves to be published in ACP since it provides a wealth of new information constraining sources of H₂. These are the first measurements of H₂, CO and other gases taken directly from the exhaust of specific combustion sources. These data fill a large void in the information available on H₂ and these data should enable a more accurate estimate of H₂ emissions using global models. This is becoming increasingly important in light of the potential use H₂ as a large scale energy carrier. The manuscript is scientifically sound and needs only minor revision before publication (see comments below).

Specific Comments: p6841, L13: replace with 'past 15 years'

L18: Yver et al, is quoted here but not in Table 1. This is inconsistent. Need to include this in the table reference if it is referred to in the text.

p6842, L18 change to 'recognising'

p6844, L19-23: This sentence is unclear and needs to be reworded and/or split up.

p6844: L14: replace 'The present' to 'This'

p6845, L24 to p6847 L22: This Instrumental part should be in a separate section in Methods and be placed before or after all the sampling site descriptions. It is confusing to the reader place here.

p6846, L7: '~2%'. It needs to be specified which gas this number applies to. If both gases, this also needs to be specified.

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p6847, L24: this should read 'at 6 waste incinerator facilities'

L26/27: Reorganise this sentence to '(...equipped to one to four boilers, they have a yearly waste throughput of 90,000-220,000 t). The incinerators are equipped with a ...'

p6848, L1: replace with '...incinerators were sampled..'

L10 replace with '...in Tedlar bags for less than...'

p6849, L22: replace with '(parts per million, 10⁻⁶)'

p6850, L2: The value of 0.05-0.2 quoted here is different to the value quoted in the table 3. Could you correct it if it is wrong or explain the difference between this value and that in the table. It would also be useful to quote the dH₂/dCO value for gas heaters in the text if this value refers to oil heaters only.

p6850: Section 3.1.2: Are residential wood heaters classed as biofuel in Table 3? If so you need to make this clear in this paragraph.

p6850-1 L25/26-L1/2: This long sentence beginning 'There are a few exception..' ending '...at the time of sampling.' is confusing and needs to be split up and reworded.

p6851, L19: reword to '...but also the H₂/CO ratios are also smaller.'

p6852, L7: '(dD ~ +130 per mil)'. This is not any of the values quoted in Table 3 as ambient air. You must quote where this value has come from.

L10: 'fuel-air ratio' is referred to in this sentence. This is very confusing as on p6851 L 24 the 'fuel-air ratio' is described in detail and then the terminology changed in the following section. Need to keep to the same terminology or else highlight that this value is the inverse of the last described fuel-air ratio.

L26-28, L1 p6852: Sentence beginning and ending ' In such a one-source.....case for our samples.' The discussion of results in relation to the Keeling plot needs to be described in further detail to make it a useful addition to the discussion.

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L28: '...specific end-member isotopic signature..' This terminology is confusing and needs to be replaced/clarified.

p6854, L2: 'The ratios from our wood-fire exhaust (2.4 and 3.9) bracket this ratio with reasonable agreement.' It needs to be clarified is these values are included in the 3.3 value for biomass.

p6854, L24 'a less pronounced imbalance'. Reword this eg. more diesel emission/vehicles than previous years.

L26: 'above mentioned 4 yr' specify the years as the sentence sounds incomplete.

p6855 L3: Replace with 'despite little change in the relative'

L5: Replace with 'are likely to be due to the'

L7: replace 'and combine these'

L18-24: These sentences are difficult to read and need to be structured differently to clearly get across the point that is being made.

p6856, L22: Remove 'While such'.

p6857, L17 L22: Replace Fig. 5 with Fig. 5a.

p6858, L24: 'there is confirmatory indication' Reword this to for example 'evidence'

p6859, L10: change to 'unlikely to be applicable'

L16: 'signatures of these exhausts' Specify which exhaust type eg. oil, gas, biofuel etc.

Table 1: Place a + sign in front of the positive isotope signatures as you have in the text.

Table 3: f: highly variable: This should be defined with a range or values in the key

Figure 1: This plot does not come out clearly when printed. The black outline of the waste incinerator needs to be removed. The grey + symbol needs to be defined in the

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key on this plot.

Figure 2: The axes and text on this plot are all too small to be read and need to be made bigger for the plot to be useable.

Figure 4: The grey + needs to be defined in the key.

Figure 5: The blue square for GFED H2 cannot be easily seen on the plot. This point needs to be changed in colour.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 6839, 2012.