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Interactive comment on “A 15 year record of high-frequency, in situ measurements of hydrogen at Mace Head, Ireland” by A. Grant et al.

Anonymous Referee #1

Received and published: 11 November 2009

Title: A 15 year record of high-frequency, in situ measurements of hydrogen at Mace Head, Ireland

Authors: A. Grant et al.

The submitted manuscript presents a 15-year dataset of continuous observations of molecular hydrogen (H₂) at the coastal measurement site Mace Head in Ireland. This data set represents one of the longest time series of continuous H₂ mixing ratios worldwide. An associated Lagrangian dispersion model analysis allows distinguishing periods representative for background conditions and episodes influenced by H₂ emissions. Thus, the data set can provide very valuable and up-to-date information about the trend of the atmospheric H₂ burden in background air as there are contradictory H₂ trend estimates reported in the literature that are partly based on rather short time

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series. When looking at the episodes with advection of polluted air masses, the Mace Head data set also makes it possible to e.g. investigate at the H₂ / CO ratios as it was recently done by several authors but in urban or suburban environments. The paper is within the scope of 'Atmospheric Chemistry and Physics' and is of sufficient originality to merit publication in this journal. The paper is scientifically sound, well written and needs only minor revisions before publication (see comments below).

Specific comments:

Page 20196, line 4: remove 'a wealth of'

Page 20196, line 15-16: '... the latitudinal gradient reflects the large photochemical source from southern latitudes ...'. How was that proofed and extracted from influence of a weaker soil sink in the southern hemisphere? Use better 'low northern latitudes' than 'southerly latitudes'

Page 20197, line 14 and line 23: 'Ehhalt' is misspelled. Check also the rest of the document.

Page 20198, lines 2-5: add the paper by Lallo et al., GRL, 2008 to the references. Mention some numbers of the in-situ studies to stress the large site-to-site differences as well as the observed variability at specific sites.

Page 20198, line 25 and line 29: remove 'on the West coast of Ireland' as it is also mentioned in the site description below.

Page 20199, sampling location: The authors only refer to the AGAGE programme. What's about all the other programmes that provide continuous data? Reactive gases, aerosols etc. What's the height of the inlet above ground?

Page 20200, lines 5-7: add the information given on page 20201, line 1 here that injections were made every 20 minutes and ambient air samples were measured every 40 min'. Add some technical details. What is the size of the sample loop? Are there any other modifications made to the default set-up the RGA3? Did you use peak height

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or peak area for the mixing ratio determination?

Page 20200, line 15-18: The authors speak about a good agreement but mention a difference of 16ppb. Mention the (known) offset of the two different scales, otherwise it is difficult to understand for the reader.

Page 20201, lines 3-5: remove 'of the possible 183,900 measurements'. Why not very simple: 'Data coverage was 79% ...'.

Page 20201, lines 22-24: 'In Fig. 2 we plot the monthly mean H₂ in Northern Hemispheric baseline air, these data display both the seasonality and variability in mole fractions over the 15 year period.' To my mind, Fig. 1 does show the variability even better. Fig. 2 mainly reveals the inter-annual variability.

Page 20201, lines 24: How were the data de-trended when there was no significant trend detected (see next page, lines 19-20)?

Page 20202, lines 1-2: To my mind, it is not correct to compare the observed annual cycles with the cycles reported by Simmonds et al. (2000) since the Simmonds data are also part of the present data set. Please reword the sentence that it becomes obvious to the reader that the present paper at the extended data set only confirms the findings of Simmonds et al. for the first 4 years of the 15-year record.

Page 20202, lines 15-17: Replace '(Fig. 3)' by 'See Fig. 3 for a comparison of the H₂ and CO seasonal cycles.' Please elaborate why the 3-month offset in spring maximum can be explained by the dominant soil sink.

Page 20203, lines 22-23: 'In Fig. 1 periods of intensely elevated H₂ above the baseline ... can be seen.' Please add the baseline, i.e. something like a running mean of the baseline values, in Fig. 1.

Page 20204, lines 17 ff.: Hard to see in the inset of Fig. 7 that the data centre at 120ppb CO. See also comment below to Fig. 7.

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Page 20205: The authors are investigating the H₂ to CO ratio during pollution events. I do agree that it is necessary to de-seasonalize both the H₂ and CO data. The authors did that by subtracting baseline values. How was this exactly done? Did they subtract monthly baseline means or did they determine a specific baseline value for each measurement point by e.g. interpolating the baseline data? After subtraction of the baseline values, why did the authors use all data (Fig. 7B also shows negative values) and not only the data above baseline as they are interested in H₂ to CO ratios from emissions? The slope might not be strongly affected as it is mainly determined by the very high data. However, the authors should consider determining their slopes with only data above baseline. How was the slope calculated? Most of the common spreadsheet calculation programs (like Excel) do only consider errors for the y-component and thus retrieve slightly incorrect slopes. The correct approach is an orthogonal or reduced major axis regression analysis.

Page 20205, line 13: what are ‘raw H₂ mole fractions’?

Page 20205, lines 13-15: An extrapolation to CO=0 makes no sense. The intercept can be dominated by the seasonal cycle of the different trace gases.

Page 20205, lines 20-23: ‘Winter data cover a broader range and show a higher correlation coefficient due to lower H₂ deposition.’ I do not agree with this statement. A higher correlation coefficient might be most likely a result of the broader range of data. If there is no variability at all, the correlation coefficient might be very low. If there is a steady soil sink that is just stronger in summer, the data are simply shifted or even not shifted in this case here because the seasonal variation is subtracted. Higher concentrations in winter can be also caused by less vertical dilution and thus a smaller buffer volume that let emissions appear more pronounced close to the ground and lead to an accumulation of freshly emitted trace gases in the lowermost levels. Comment: Fig. 7 A and B show that the consideration of the seasonality of the background concentrations leads to a better H₂ to CO correlation.

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Page 20207, line 16 and page 20208, line 5: ‘The conversion value for non-transport emissions ... a range of values from 0.07 to 0.57 has been used ...’. ‘The best match is for a non-transport scenario of 0.06 ...’. It is hard to understand what these numbers mean without having a look at Fig. 8. Is it ‘The best match is for a non-transport molar H₂ to CO ratio of 0.06 ...’?

Page 20209: The authors discuss if the observed H₂ to CO ratio has to be corrected for H₂ soil deposition? What’s about the CO soil deposition? The best guesses of soil deposition velocities in the literature are pretty similar for H₂ and CO.

Conclusions: The authors mention at the very end of the introduction that ‘this 15 year dataset may provide essential information to improve assessments of the effect of a possible future H₂ economy on atmospheric composition’. No reference is made to this statement in the conclusions. The authors should refer to that and should discuss how the assessment of the effects of a future H₂ economy on the environment can profit from the present study.

References: Simmonds (2009) ??? Is it published?

Fig. 1: add a curve that represents mean baseline values.

Fig. 4: add horizontal line at $y = 0$

Fig. 5: are these the baseline data that are subtracted from the data set?

Fig. 6: shift the two time series a bit to improve readability.

Fig. 7: improve quality of Fig. 7. The inset is hard to read. Why not three Figs. A, B, and C?

Fig. 7A: mark the episode mentioned on page 20204-20205 with an arrow. Add a second arrow that indicates the circular direction with time (counter clockwise?). Use colors, the different symbols are hard to distinguish. What is really shown in Fig. 7A? Are these daily averages? Please clarify. Fig. 7B: add regression coefficients.

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Fig. 9: the authors state that episodes with southerly advection are pretty seldom at Mace Head (250 in total). I would be curious to see the number of data points that are considered for the three seasonal cycles (southerly, European, baseline) for each month (e.g. on top of the graph?). This can give valuable information to the reader about the representativeness of the mean values. Error bars illustrating standard deviations of the mean might also be helpful.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 20195, 2009.

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