Interactive comment on “Sources and sinks of acetone, methanol, and acetaldehyde in North Atlantic air” by A. C. Lewis et al.

Anonymous Referee #1

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Review - "Sources and sinks of acetone, methanol and acetaldehyde in North Atlantic air", by Lewis et al.

This paper presents a set of field measurements taken in the marine boundary layer on the West coast of Ireland. The focus here is on the oxygenated compounds, methanol, acetone and acetaldehyde. The authors make the point that, according to their measurements, these compounds dominate both the mass and HO radical sink for non-methane hydrocarbons. The data are compared to an explicit box model calculation to further elucidate the sources and sinks of these compounds. Precursor compounds for each species are identified. The paper brings in several insights into oxygenate chemistry, the relatively small number of precursors, the apparent lack of ocean sink
for acetone and acetaldehyde and the inexplicable acetaldehyde variability and concentration. The paper is interesting, well organised and clearly presented. However, prior to acceptance in ACP the following points should be addressed to further improve the paper.

1) Since this paper addresses processes in the marine boundary layer it seems appropriate to alter the paper's title to include the word "marine" after "Atlantic".

2) The paper needs to be better referenced. Several relevant works are not mentioned. For example, in the first line of the introduction (line 20) it seems fitting to cite the pioneering work of Singh et al 2001 (Nature, 410). Data from over the Pacific ocean MBL contained in this paper can also be included in Table 1. Recent measurements of acetone and methanol also made over the Atlantic (Williams et al. 2004 GRL) are similarly overlooked. See also points 3 and 9 in this regard. The reference e.g. Galbally et al 2002 (line 26) should be Galbally and Kirstine 2002.

3) In the introduction, it would be helpful to briefly summarise for the three oxygenates considered, the present understanding of the oceans role in their global budgets.

4) Methodology - since there are several measurement issues for oxygenated species (see point 9) it is important that all sampling information is given. The 25m sampling manifold description should include, the material used and the approximate residence time. Although the work of Hopkins et al is referred to, details of the accuracy and precision of the measurement method should be given (at least for the oxygenates).

5) Page 1290, line 3. Figure 3 is referred to but the authors appear to mean Figure 2.

6) Section 3.2. p1290, line 12. The authors must be more specific here. How do the oxygenates "dominate?" Are methane and CO included in this perspective? Although this is explained later, it is not clear at this point.

7) p1290, line 27. "is" should be "are" since sinks is plural.

8) The authors may wish to compare their variability analysis to that of Williams et al...
2000 JGR 105, D16, 20473 who also made this analysis for measurements of oxygenates in Atlantic air. Interestingly the aforementioned paper also recorded a lower than expected variability for acetaldehyde, while methanol and acetone were close to the variability/lifetime trend defined by other species.

9) Clear from the modelling section figure 10, is that the levels of acetaldehyde measured (average 440 pptv, max 2.12 cannot be reproduced by the model. Several recent works indicate the possibility of artifacts in the measurement of acetaldehyde, and these artifacts appear to be independent of the instrument used. Singh et al. 2004 JGR vol 109 indicate that an interference was found in stratospheric measurements with an airborne GC system. Northway et al. 2004 (Atmos. Env 38, 6017) show that interferences also have been seen in stratospherically influenced air for a CIMS type instrument, and that a variety heterogeneous inlet reactions can produce acetaldehyde. Furthermore Apel et al. 2003 JGR 108, D20 also mentions acetaldehyde production on surfaces as a problem in their GC-MS system, a system very similar to the one used here. With this literature in mind it seems reasonable that the authors should consider the possibility that the acetaldehyde data may be artificially high in the text. If specific tests have been carried out to eliminate this concern (e.g. ozone at inlet) they should be mentioned. An instrumental source of acetaldehyde (a constant artifact production) would also be a valid explanation for the low observed variability of this species.

10) An interesting addition to this paper would be formaldehyde or PAN data if available. This would help judge the validity of the acetaldehyde data. The presumably wider range of precursors for formaldehyde would provide an interesting counterpoint to the three oxygenates discussed.

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