

Interactive comment on “Global atmospheric budget of acetaldehyde: 3-D model analysis and constraints from in-situ and satellite observations” by D. B. Millet et al.

Anonymous Referee #1

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General comments:

This paper constructs a global budget for atmospheric acetaldehyde using the global chemical transport model GEOS-CHEM, aided by satellite and aircraft observations to constrain its uncertainty. Atmospheric acetaldehyde is mainly produced photochemically by the oxidation of alkanes, alkenes, isoprenes, and ethanol. It is also emitted by biogenic, anthropogenic, and biomass burning activities, as well as by photodegradation of CDOM in oceans. In particular, to constrain the ocean source, the authors used satellite observations of color dissolved matter and empirical relations derived from field observations to construct the global air-sea flux. The resulting global net flux

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from sea to air is 58 Tg/a, much smaller than the 125 Tg/a previously derived by Singh et al. [2004] based aircraft measurements in the marine boundary layer.

The authors went on to validate their budget by comparing simulated acetaldehyde concentrations to available aircraft measurements over global land and oceans. Over land, both the concentrations and vertical gradients of simulated acetaldehyde agree well with observations, except in areas heavily impacted by urban outflow. The authors hypothesize that an important photochemical source is missing in the current model chemical mechanisms. Over the oceans, simulated acetaldehyde concentrations generally agree with observations in the MBL; but measurements show a persistent 100–400 ppt background in the marine free troposphere not captured by the model. The authors also looked at the PAN:NO_x ratio over land and ocean as additional validation of the acetaldehyde budget. They concluded that the simulated acetaldehyde is consistent with the observed PAN:NO_x ratio both over land and ocean. They stated that there is no strong evidence for a large oceanic acetaldehyde flux as derived by Singh et al. [2004], but no definitive explanation was offered as to why the measured values are high in the marine free troposphere (except citing Northway et al., 2004). Lastly, the authors estimated the relative importance of the background (i.e., current sources) ethanol and acetaldehyde sources if the U.S. were to adopt ethanol fuel.

The topic of this paper is important and within the scope of ACP. The methodology is novel and sound. The manuscript is well written. I recommend publication after minor revision to address the following specific issues:

Specific comments:

1. Page 24247, lines 20–25; and Figure 9 and 10: The authors stated that the current model produces a good PAN:NO_x ratio over the ocean surface, and use that as the main argument against a large sea-to-air acetaldehyde flux. It would be useful to see what the acetaldehyde profiles and PAN:NO_x profiles look like when the 125 Tg a⁻¹ ocean source is used.

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2. Table 2: Units are in Tg y⁻¹. Everywhere else in the main text the budget is in units of Tg a⁻¹. Modify to be consistent.

3. Figure S2 is confusing. Why is the emission factor for acetaldehyde and ethanol the same? If this is due to a lack of speciated emission factor for OVOCs in the MEGAN inventory, which resulted in the same biogenic emission estimates for acetaldehyde and ethanol, then this should be pointed out in the main text. The current text gives the impression that there is speciated information in MEGAN. Also, what exactly is plotted in Figure S2? Is it one of the emission factors ϵ_i , or is it the weighted $\sum \epsilon_i \chi_i$, $i = 1,5$?

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 24225, 2009.