

# 1 Global atmospheric budget of acetaldehyde: 3D model analysis and 2 constraints from in-situ and satellite observations

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## 32 Supplemental Information

### 33 34 1 MEGANv2.1 details

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36 The MEGANv2.1 acetaldehyde emission parameterizations are primarily based on whole-canopy  
37 flux measurements. The exceptions are the light dependence and soil moisture response, which  
38 are based on enclosure measurements due to a lack of canopy-scale observations. Surface-  
39 atmosphere exchange of acetaldehyde has been measured using tower-based flux systems  
40 deployed in ten ecosystems including tropical forest (Karl et al., 2004; Karl et al., 2007), warm  
41 conifer forest (Karl et al., 2005), cool temperate conifer forest (Schade and Goldstein, 2001; Karl  
42 et al., 2002), temperate broadleaf forest and plantation (Karl et al., 2003; Jardine et al., 2008),  
43 boreal forest (Rinne et al., 2007), and cropland/grassland (Warneke et al., 2002; Schade and  
44 Custer, 2004). Nearly all of these studies used proton-transfer reaction mass spectrometry (PTR-  
45 MS) and the eddy covariance, or disjunct eddy covariance, approach (see Karl et al., 2002). The  
46 only exception is Schade and Goldstein (2001) who used an automated relaxed eddy

47 accumulation system with on-line gas chromatography/flame ionization detector (GC-FID). The  
48 daytime fluxes observed at these sites for warm, sunny conditions range from no emission (or a  
49 net uptake) to an emission of about  $350 \mu\text{g m}^{-2} \text{h}^{-1}$ . The observed temperature response factor ( $\beta$ )  
50 ranges from 0.05 to 0.19. The fluxes for the two cropland/grassland sites are both on the lower  
51 end of the emission range, but conifer and broadleaf forests cover the entire range of reported  
52 emissions and so are grouped together. The MEGANv2.1 acetaldehyde emission factors are  
53 based on the median values from the above studies:  $200 \mu\text{g m}^{-2} \text{h}^{-1}$  for forests and  $20 \mu\text{g m}^{-2} \text{h}^{-1}$   
54 for herbaceous landscapes. The median temperature response ( $\beta = 0.13$ ) reported for these  
55 studies is used for all ecosystems. The MEGANv2.1 LDF and LAI dependence is based on the  
56 Jardine et al. (2008) analysis of leaf level observations and a comparison of above-canopy fluxes  
57 at three temperate forests. The leaf level acetaldehyde emissions reported by Holzinger et al.  
58 (2000), Kreuzwieser et al. (2000), and Rottenberger et al. (2008) provided the basis for the  
59 MEGANv2.1 soil moisture response algorithm.

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61 Due to the marginal ability of PTR-MS systems to quantify ethanol, there are relatively few  
62 observations of ethanol emissions from vegetation. Of the ten above-canopy studies used to  
63 characterize acetaldehyde fluxes, only one study (Schade and Goldstein, 2001) reports ethanol  
64 emissions. They observed ethanol emissions that were about a factor of 2 higher than  
65 acetaldehyde. The temperature response for ethanol at this site was similar to that observed for  
66 acetaldehyde. Additional field measurements of ethanol emissions (Fukui and Doskey, 1998;  
67 Kirstine et al., 1998) have used enclosure techniques and also observed ethanol emissions that  
68 are somewhat higher but of the same magnitude as acetaldehyde fluxes. As a starting point for  
69 introducing ethanol emissions in MEGANv2.1, we have used the parameterization developed for  
70 acetaldehyde to also represent ethanol emissions. As a result, these ethanol emission estimates  
71 are relatively uncertain; more observations of ethanol fluxes are needed to improve this initial  
72 approach.

## 73 74 **2 Air-sea flux calculation**

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76 We compute the flux of acetaldehyde across the air-sea interface using the Liss and Slater (1974)  
77 two-layer model:

$$78 \quad F = K_L \left( \frac{C_G}{H^*} - C_L \right), \quad (1)$$

79 where the flux  $F$  is determined by the acetaldehyde concentrations in the gas and liquid phase  
80 ( $C_G$ ,  $C_L$ ), the total transfer resistance ( $1/K_L$ ), and the dimensionless Henry's law coefficient for  
81 acetaldehyde  $H^*$ . In the aqueous phase acetaldehyde is partially present in the hydrated form  
82  $\text{CH}_3\text{CH}(\text{OH})_2$ , so that  $H^*$  is actually the apparent Henry's law coefficient defined as the  
83 equilibrium ratio of the concentration in air to the sum of hydrated plus unhydrated aldehyde in  
84 the aqueous phase (Betterton and Hoffmann, 1988). The overall transfer resistance is the sum of  
85 those in the individual phases,

$$86 \quad \frac{1}{K_L} = \frac{1}{\alpha k_l} + \frac{1}{H^* k_g}. \quad (2)$$

87 We derive  $k_l$  and  $k_g$  using the wind-speed dependent parameterizations of Nightingale et al.  
88 (2000) and Asher (1997), respectively. The enhancement factor  $\alpha$  reflects the extent to which  
89 acetaldehyde hydration increases the rate of aqueous diffusion; here  $\alpha = 2.0$  following Zhou and  
90 Mopper (1997).

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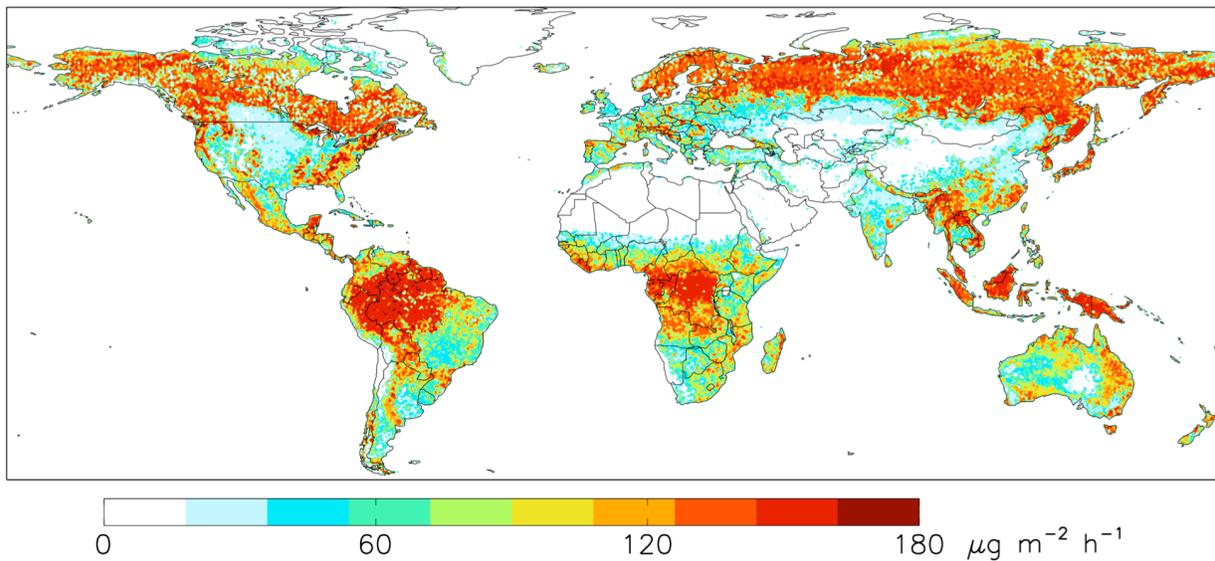
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161 **Fig. S1.** Global weighted emission factor distribution ( $\sum \varepsilon_i \chi_i$ , see Equation 1 of the main text) for  
162 acetaldehyde and ethanol used in MEGANv2.1, gridded to  $0.5^\circ \times 0.5^\circ$ . Biogenic emissions of the  
163 two compounds are computed using the same vegetation-specific emission factors  $\varepsilon_i$  as described  
164 in SI Section 1 above.

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