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**
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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 μg m^{-2} h^{-1}. The observed temperature response factor (β)
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 μg m^{-2} h^{-1} for forests and 20 μg m^{-2} 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.
60
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 $$F = K_L\left(\frac{C_G}{H^*} - C_L\right), \quad (1)$$

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 $CH_3CH(OH)_2$, 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 $$\frac{1}{K_L} = \frac{1}{\alpha k_l} + \frac{1}{H^* k_g}. \quad (2)$$

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 α 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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Photochemical Production — 0.3 Tg
Biogenic — 22. Tg
Anthropogenic — 2. Tg
Biomass Burning — 0.07 Tg
Photochemical Loss — 19. Tg
Dry + Wet Deposition — 5. Tg

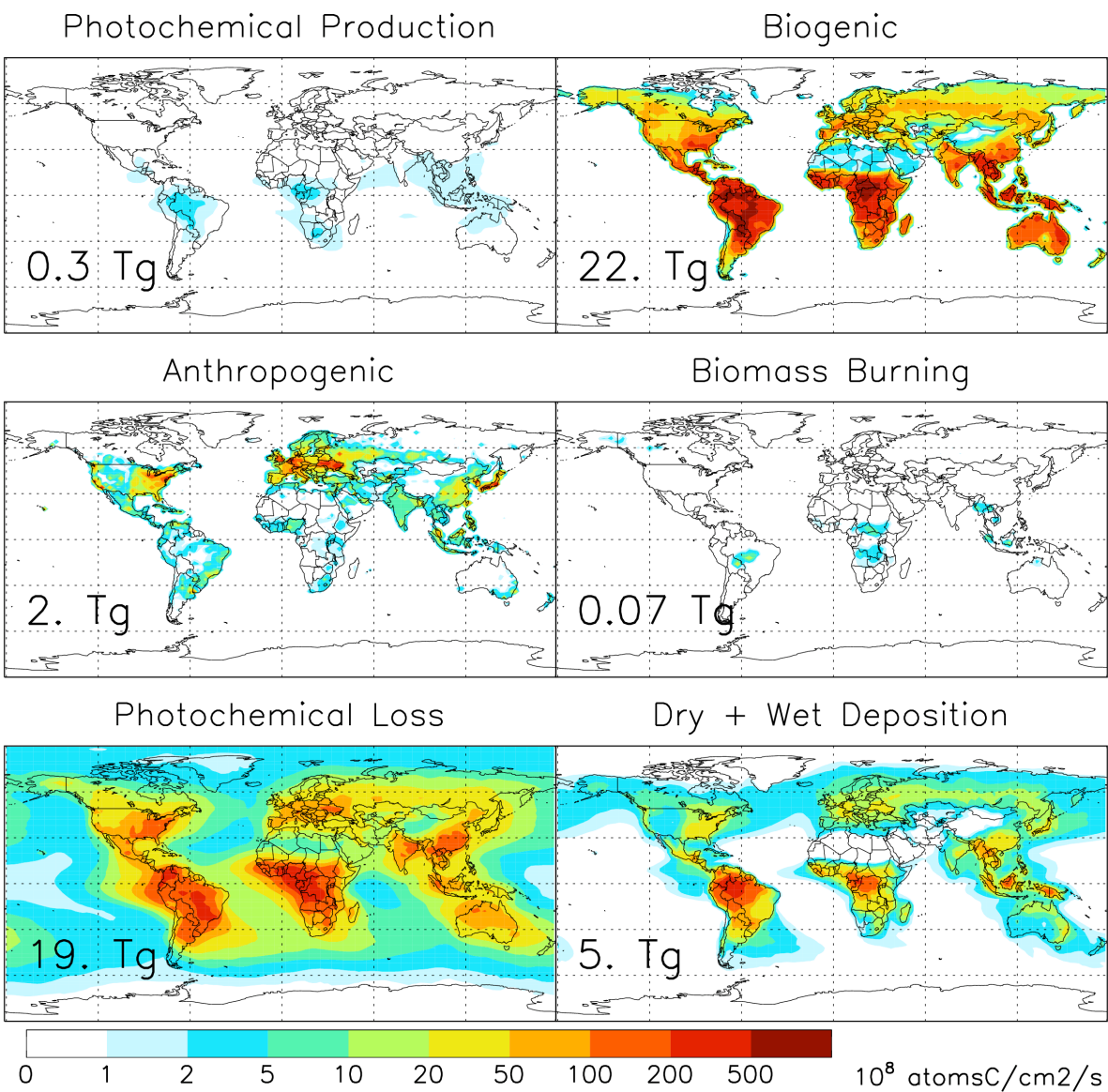0   1   2   5   10   20   50   100   200   500 10^8 atomsC/cm2/s

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Fig. S1. Annual average sources and sinks of ethanol in GEOS-Chem. Shown are photochemical production, biogenic emissions from live and decaying plants, anthropogenic emissions (urban/industrial + biofuel), biomass burning emissions, photochemical loss, and deposition.
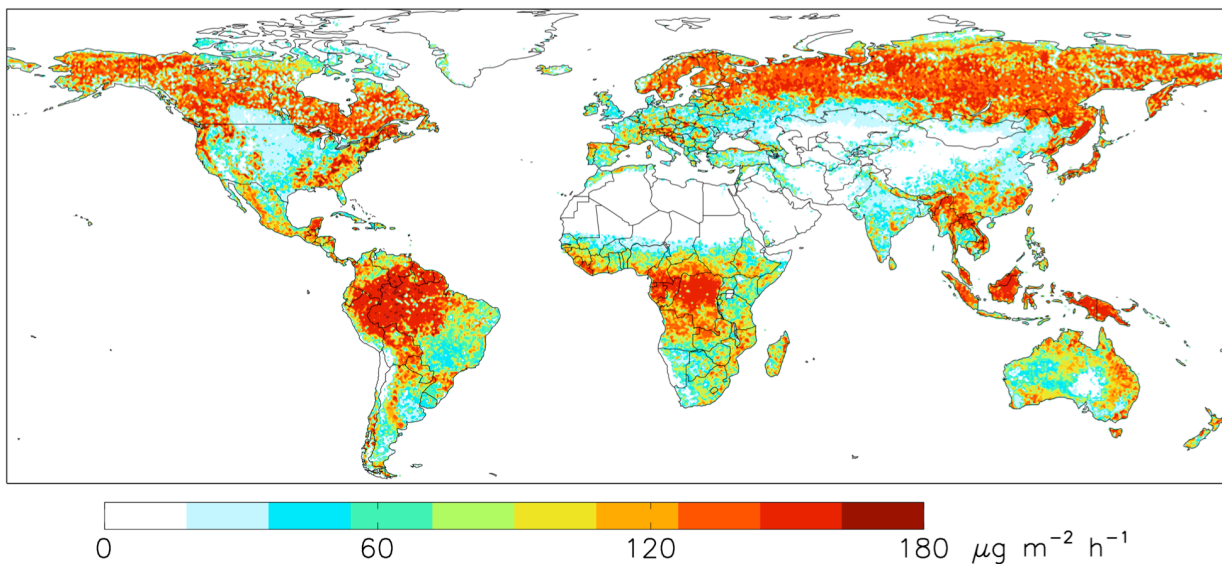
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164 **Fig. S2.** Global emission factor distribution (gridded to 0.5° × 0.5°) for acetaldehyde and ethanol
165 used in MEGANv2.1.

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