

Interactive comment on “Plant physiological and environmental controls over the exchange of acetaldehyde between forest canopies and the atmosphere” by K. Jardine et al.

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We are very thankful for the review by Dr. Rosenstiel which should help start a public discussion and encourage the use of our stable carbon isotope technique in future studies aimed at investigating the role of plant physiology on the exchange of oxygenated VOCs between plants and the atmosphere. Our responses are below:

Reviewer comment: The use of the PTR-MS to estimate transpiration is creative, but as a non PTR-MS expert it would be useful if the authors could show (or cite) some data confirming that this MS based approach to estimating transpiration does, in fact, scale with more traditional (IRGA/porometer) techniques, especially since this is a key aspect of their study.

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Response: Ammann et al. (2006) used a PTR-MS to measure water vapor fluxes in comparison to a common reference system consisting of an infra-red gas analyzer (IRGA). As with the current study, water vapor was detected with the PTR-MS at m/z 37. The water vapor flux obtained with the PTR-MS showed a very good agreement with the flux of the reference system. We propose to insert this reference on Page 2693, Line 5.

Reviewer comment: I found the kinetic isotope effect compelling, and perhaps the strongest evidence for selective acetaldehyde uptake mediated by stomata. However, with the data presented I'm wondering why the authors believe stomatal resistance alone is sufficient to account for the 5ppm differences observed in the intact poplar branches, especially considering the wide-open nature of poplar stomata. The authors should expand their discussion of this point, particularly with regards to some of the current thinking about the extent to which stomatal resistances do/do not contribute (and how much) to fractionation of other gaseous compounds (particularly CO₂). I realize the authors would like to conclude there is no deposition to leaf (branch) surfaces in their experiments (and likely this is very small), but the argument given that 15h of continuous fumigation should saturate surface binding is really speculation at best, especially since the total leaf /branch surface area was not presented nor do we really know what the capacity for deposition might be. This could be particularly problematic if leaf microorganisms (bacteria/fungi/etc?) are present (which they would be) that might also uptake and utilize acetaldehyde over these time scales. Ideally, in this study the authors would have incorporated an ABA-feeding experiment. Hormonally regulating stomatal aperture (closing) while simultaneously maintaining leaf cell wall/epidermis hydration status would be the real test for of the surface deposition hypothesis and also would nicely clarify the fractionation results. I hope someone in the future integrates ABA based experiments with GC-IRMS approaches.

Response: The calculated KIE associated with the uptake of acetaldehyde by poplar branches (5.1 +/- 0.3 per mil) is close to the theoretical KIE occurring due to diffusion

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in air (4.4 per mil). Therefore, we can conclude that fractionation during diffusion in air is the most important process that determines the overall KIE associated with the uptake of acetaldehyde by the poplar branches. In analogy with what is known about carbon dioxide during photosynthesis, if stomatal uptake is the dominate pathway for acetaldehyde exchange then the KIE during the diffusion of acetaldehyde into leaves can be described by $KIE = (1 - C_o/C_a)a$, where C_a is the ambient concentration of CO_2 ($g\ m^{-3}$) and C_o is the intercellular concentration of CO_2 ($g\ m^{-3}$) at the site of oxidation, and a is the fractionation occurring due to diffusion in air (Farquhar et al., 1989). During net uptake, increasing stomatal resistance should tend to decrease C_o/C_a while decreasing stomatal resistance should tend to increase it. Therefore, as observed in this study, the wide-open nature of poplar stomata should allow for the full value of a to be expressed in the observed KIE. In contrast, conditions favoring more closed stomata during net uptake should decrease the KIE by reducing the importance of a . Future work using ABA to induce stomatal closing should attempt to verify this important prediction if stomatal uptake is indeed the dominant exchange mechanism between plants and the atmosphere. Ideally, future studies investigating the role of stomatal versus surface exchange of acetaldehyde would investigate the influence of stomatal resistance on both the KIE associated with uptake of acetaldehyde and the acetaldehyde exchange velocity.

Reviewer comment: Finally, the authors seem to want to drive/relate acetaldehyde exchange dynamics to solar radiation (i.e. discussion of sun shade leaves, canopy density, self shading, etc) suggesting in numerous places that emission is a function of light. Of course biochemically there may be some relationship to light (light enhanced rates of mitochondrial respiration?), but in the data presented the authors can't really separate light-enhanced emission from light regulated control over stomatal aperture. This, of course, is very different from light-dependent VOCs (such as isoprene) where the biosynthesis can be directly related to PAR and electron transport. The enhanced emission from sun leaves in the canopy may simply reflect lowered stomatal resistance in concert with higher transpiration rates. Although one could relate G_s to PAR in

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some systems and under some conditions, in general I don't think the statement "acetaldehyde compensation point is a function of light" is really what the authors intend to suggest. A subtle point, but critical if we are all to agree on the physiological parameters that ultimately regulate acetaldehyde emission and the environmental drivers that matter.

Response: We propose to clarify this point by inserting the following paragraph on Page 2661, Line 6.

In this paper, we suggest that stomatal/mesophyll resistances influence the acetaldehyde exchange velocity (the slopes of the compensation point curves) while light/temperature influences the acetaldehyde compensation points (x-intercepts of the compensation point curves). Consequently, stomatal resistance does not affect the acetaldehyde compensation point but rather its exchange rate with the atmosphere. Therefore a small stomatal resistance value leads to a high exchange velocity with high emission rates at ambient concentrations below the compensation point and high uptake rates at ambient concentrations above the compensation point. Our results are supported by a complimentary study which found that species dependent differences in acetaldehyde exchange velocities were largely attributed with differences in stomatal resistance (Rottenberger et al., 2008). The direction of the net exchange (net emission or net uptake) can only be determined by the compensation point and ambient concentrations. The net emission rates of sun leaves in the canopy must therefore be caused by an increase in compensation points over the ambient acetaldehyde concentrations as was observed in the branch enclosure studies in the light. Because temperature also increased in the light, we were not able separate light and temperature effects on the compensation point. While temperature has been clearly shown to increase compensation points (Karl et al., 2005), Jardine (2008) has proposed a plausible biochemical mechanism for light enhanced acetaldehyde production. It is now widely accepted that day respiration rates are suppressed relative the night respiration in leaves. However, there is a continuous export of triose phosphates from chloroplasts

during photosynthesis. This may lead to enhanced pyruvate concentrations in the cytosol which may then stimulate ethanolic fermentation rates. This same mechanism has been proposed for the transient bursts in acetaldehyde emissions during light to dark transitions (Karl et al., 2002a) termed the pyruvate overflow mechanism. However, Jardine (2008) extended this view by observing bursts of acetaldehyde as well as carbon dioxide, ethanol, and acetic acid following similar light to dark transitions. Therefore, this process was termed light enhanced dark fermentation (LEDF) due to its strong analogies with the well known, but poorly understood light enhanced dark respiration process (LEDR). The role of light on the compensation point of acetaldehyde and the LEDF and LEDR process deserves additional research.

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