

## ***Interactive comment on “Environmental controls over methanol emission from leaves” by P. Harley et al.***

### **Anonymous Referee #1**

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#### GENERAL COMMENTS

The paper presents a large amount of measured data of leaf-level methanol emission and stomatal conductance from different plant species under artificially varied light and temperature conditions. The authors successfully modelled the observed time series of methanol emission by using a combination of the Niinemets and Reichstein (2003) approach to account for buffering in the liquid leaf pool, and a methanol production rate that is parameterised as an exponential function of leaf temperature. The latter function was fitted to individual datasets of the different plant species resulting in individual values of the standard methanol production rate at 30degC and the corresponding exponential coefficient. The integral methanol emission of the individual experiments was also compared to the corresponding carbon assimilation.

The present work is an important step in the development of a useful methanol emission algorithm that can be applied in large scale models and certainly deserves publication in this journal. Unfortunately, the manuscript has some shortcomings concerning its structure that affect the clearness and the readability and also make it lengthy. The 'Discussion' section looks like a paper within the paper. It mainly treats the model simulations and includes its own introduction, methods, results, and discussion part. However, the model simulations represent an integral part of the manuscript and should not be introduced only in the 'Discussion' section. In fact, model results are displayed in most Figures that are presented already much earlier in the paper. In particular, the entire introduction and method description of the modelling exercise in Chapter 4 (from p2607, line 19 to p2611, line 16) should be moved to Chapter 2 and could be shortened considerably.

One main result of the paper is that (apart from short term variations captured by the Niinemets and Reichstein approach) methanol emission rates are mainly controlled by the leaf temperature. It would be therefore very useful to collect the respective results (Pm30 and beta values) for the different plant species/experiments in a table instead of only present them scattered in the various figure captions.

#### SPECIFIC COMMENTS

p2597, line 16: The statement "...Niinemets and Reichstein (2003a) ... developed a general model of VOC emission from plants..." is misleading and contradicts the important limitations of this approach (need of prescribed production rate) mentioned in the following paragraph.

p2596, line 8ff: Fukui and Doskey (1998) and Kirstine et al. (1998) did not use relaxed eddy accumulation or eddy correlation techniques but they applied static enclosure systems for the methanol flux measurements.

p2598, line 1f: The authors state that the major goal of this paper is to establish the influence of different controlling parameters on the methanol emission under "naturally

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varying conditions of light and temperature". I think this is a bit of an overstatement considering the often arbitrary (no coherent 24h cycles) and partly large step-like changes of light and temperature in the presented laboratory experiments.

p2598, line 24ff: Which environmental conditions were experienced by the whole plant/sapling, apart from the enclosed leave, during the measurements? Were they always constant? It would be interesting to discuss, whether or not the results of the present paper indicate that the methanol emission is only depending on local leaf conditions.

p2598, line 26: All presented emission measurements were obtained with zero air entering the leaf cuvette. Please shortly discuss the possible implications of using zero air instead of typical ambient methanol concentration concerning simultaneous deposition processes or a compensation point. I don't think that this issue is critical but it deserves at least a short consideration.

p2599, line 9: If I understand the method description right, only the air exiting the cuvette was analysed for CO<sub>2</sub> and H<sub>2</sub>O, while the concentrations in the entering air was calculated from the gas mixing flow rates!? Whether this is the case or not, the uncertainty of H<sub>2</sub>O flux measurements should be quantified, because it has a crucial effect on the calculation of the stomatal conductance and thus on the modelled methanol emission under dark or low light conditions (where least agreement between measurements and model simulations was found).

p2604, line 16: The observation, that methanol shows a better linear relationship with transpiration than with stomatal conductance deserves further discussion. It may indicate that the assumption of Niinemets and Reichstein (2003) considering only diffusive transport of methanol within the leaf is not fully adequate. Since transpiration implies a major 'advective' transport of water (including dissolved compounds) within the leaves and the entire plant, it may also contribute to the control of methanol release to the atmosphere (especially of methanol that is not produced near the stomata, but in other

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parts of the plant like e.g. the roots).

p2606, line 16ff: This statement about planned future activities should be moved to the end of the 'Conclusions' section.

p2608, line 3ff: This statement is misleading, because the pools do not stay in equilibrium, when  $g_s$  is changed (as described later in the paragraph). The statement could be formulated more clearly as follows: "...in steady-state equilibrium conditions, methanol emission equals the production rate and is independent of stomatal conductance."

p2617, line 17f: Not the re-equilibration of the (very small) gas-phase pool but the re-equilibration of the aqueous pool produces a time lag. So the sentence should be reformulated e.g. as follows: "...and the lag time introduced for its re-equilibration partly uncouple the instantaneous emission rate from the corresponding rate of production."

#### TECHNICAL CORRECTIONS

Figs. 1,2,10,11,13,14 are too small and/or the plotted lines are too broad.

Figs. 3,5,7: Explain the meaning of the colour shadings in the respective figure captions (not only in the captions of the succeeding figures).

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