

We thank Dr. Hörtnagl for his careful reading of the manuscript and points raised. Please find below a point-by-point reply to your comments (italic: reviewer comments).

P24007, line 8: It would be important to know more about the line heating. Methanol is known to easily stick to tube walls, therefore more knowledge about the (various?) heating temperatures could be an important information. "...the line was slightly heated above ambient temperature." seems too vague.

We will change the sentence: "...the line was slightly heated above ambient temperature." by "The sampling line was wrapped with two heating cables (20 W/m) and three thermistors were placed along the line to monitor the heating. The output of the thermistors showed that the line was on average 12°C above ambient temperature."

P24008, line 8: One measurement cycle was 2 s (maybe slightly longer as data had to be stored?), therefore I assume that about 900 data points were used in the flux calculations for each half hour. Provided the lag time was about 15 s another 7 to 8 data points would be lost due to the lag removal. Is this about right? I think the number of data points per half hour that have been used for flux calculations should be mentioned here. Also, I would like to know how many half-hourly flux values have passed all subsequent quality tests and were used for further analyses (see also next comment).

The number of data points per half hour that have been used for flux calculations is about 790 in 2009 and 605 in 2010 (due to the addition of two other masses in the measurement cycle). We will add these numbers into the manuscripts.

P24008, line 10: More information on the flux calculations is needed. Although the authors refer to the previously published Laffineur et al. (2011), a sentence or two regarding lag time determination, lag time window and also quality controls on fluxes (e.g. stationarity, coordinate rotation...) would be helpful. Regarding the lag time: in the previously published Laffineur et al. (2011) the mean time lag found (for all VOCs?) was 14.8 s, while the theoretical time lag was slightly lower (12.9 s). Lag was searched in a specific time window and, if the maximum of the covariance function was not found between 10-18 s, the lag was set to the theoretical time lag. Was this also the case for methanol? If so, it should be mentioned in this manuscript. Also, it would be interesting to know why the theoretical and not the mean time lag was used in case no clear maximum covariance was found.

We will add these sentences: "The time lag between the sonic anemometer measurements and the PTR-MS measurements was computed for each half-hour by shifting one-time series relative to the other until the absolute maximum covariance between the two-time series was determined. We used the filled-time series as proposed by Spirig et al. (2005) and described above to determine the time lag (but not to compute fluxes). This approach allowed an easier time lag determination and is similar to the averaging approach proposed by Taipale et al. (2010). The mean

time lag found using this method was 14.8 s for methanol and others BVOCs, close to 12.9 s, the theoretical value computed from the flow rate and the inlet line volume. This experimental mean time lag was used as the default value when we didn't found a maximum in the covariance function inside the [10 s, 18 s] time window. Methanol fluxes were computed using block average over 30 min periods, and 2D rotation was applied. Stationarity test (Foken and Wichura, 1996) was not applied in this study as in Brunner et al. (2007), because fluxes would hardly pass the test (more than 40% of data would have been rejected) and because this filtering did not increase the quality of our methanol data. A filter linked to anthropogenic influence (Sect. 2.4) and a stability filter (Sect. 4.3.1) were applied. Over the course of the two measurements campaigns 10138 half-hourly fluxes for methanol were recorded, of which 5481 passed all filtering criteria.

High frequency losses due mainly to the damping of concentration fluctuations in the sampling line were corrected experimentally following the method reported by Aubinet et al. (2001) using a transfer function determined by a comparison of the sensible heat flux co-spectra and the m/z 69 flux co-spectra. From this unique transfer function, a correction factor was deduced which was applied to the BVOC fluxes. For example, for a wind speed of 3 m s^{-1} (mean value of our dataset), we obtained a correction factor of 1.49."

We will change the sentence (P24016, line 6): "(26 % of the dataset)" by "(11% of the dataset after the anthropogenic filtering)".

P24008, line 20, 2.4 Data filtering: One of the crucial points that should be elaborated in detail is the data filtering and I think it is important to provide more information on methanol specific criteria. In section 2.4 the authors write that measurements from the main wind direction could be contaminated by the activities of a wood panel factory situated 3 km from the tower and point out that wood panel production is known to emit high levels of monoterpenes and methanol (Nicholson, 2003). The authors refer to a previously published paper describing isoprene and monoterpene emissions from the same site for data filtering (Laffineur et al., 2011). Fig. 2 in Laffineur et al. (2011) shows a peak of monoterpene emission ratios in the wind section of the wood factory, which is also the main wind direction. This shows that there indeed seems to be an influence of anthropogenic activities on the measurements. In order to increase the trustworthiness of the presented results, it seems important to me to know more about the quality controls used for methanol fluxes. I therefore recommend including a figure like Fig. 2 in Laffineur et al. (2011) for methanol mixing ratios before and after the data filtering which would be of great help in assessing the possibility of human activities on the measurements. The authors described a filtering criterion based on wind direction only (which would be plausible) as too restrictive and therefore used a quality criterion based on the variance of the monoterpene mixing ratios. With the factory running, this variance was very high for the main wind direction, while it was low when the factory was closed. Therefore, half hours with high variance of monoterpene mixing ratios were excluded from the methanol analysis. One of the flaws of these assumption is the possibility of keeping half hours with constant (i.e. variance is low) high monoterpene emissions due to factory production. I would therefore ask the authors to elaborate more on the possibility of having measurements influenced by the factory in their analysis. To what extent can anthropogenic influences be excluded after applying the

filter described above? This may be an important question regarding the scope of this publication, as the observed uptake could also be site-specific which could also explain why other sites have not observed similar behaviour. However, even if the methanol sink turned out to be site-specific, it should not distract from the fact that the methanol uptake in itself is an important and interesting observation in understanding biochemical life cycles of plant volatiles. After the exclusion of data with suspected anthropogenic influence the data from the “contaminated” wind sector should yield results very similar to those obtained from “non-contaminated” wind sectors. If the results differ vastly from each other it is possibly due to anthropogenic artifacts in the main wind direction. More information on this issue would be interesting. In order to be able to apply the findings of this study to other forest ecosystems the clarifications described above are of great importance.

It is easier to define a threshold on the monoterpenes variance than on the methanol variance (see figure A) to exclude precisely the data affected strongly by factory emissions. Generally, high monoterpenes mixing ratio was associated with high methanol mixing ratio. The figure B shows the effect of the monoterpenes variance filtering on the methanol mixing ratio. The monoterpenes variance seems to be a robust criterion to exclude methanol data affected by the factory. The filtering suppresses also data points outside the factory direction but in a small number of cases in comparison with the number of data point that succeed the test.

We will add the figure B to the manuscript together with these sentences: “Indeed, it is easier to define a threshold on the monoterpenes variance than on the methanol variance to exclude precisely the data affected strongly by factory emissions (27 % of data 2009-2010 was rejected). Figure 1 shows the effect of the monoterpenes variance filtering on the methanol mixing ratio. The monoterpenes variance seems to be a sufficiently robust criterion to exclude methanol data affected by the factory. The filtering suppresses also data points outside the factory direction but in a small number of cases in comparison with the number of data point that succeed the test.

It is unlikely that monoterpenes production maintains a constant high monoterpenes mixing ratio. At the chimney outlets (two main chimneys = punctual sources) located 3km away from the tower, the potential constant hot smoke flow is partially mixed with the ambient air and induces high fluctuations in the monoterpenes mixing ratio.

After the exclusion of data with suspected anthropogenic influence the data from the “contaminated” wind sector are very similar to those obtained from “non-contaminated” wind sectors as illustrated by the figure C (we have excluded the rain periods). There are no anthropogenic artifacts in the main wind direction after filtering.

P24010, line 16 onwards: With the methanol adsorption/desorption model consisting of two components (ad-/desorption in water films and degradation in aqueous-phase) it would be interesting to link it with a model describing methanol exchange during (relatively) dry environmental conditions.

The objective of our study was to explain why our site behaves as a methanol sink in contrast to what has been found at others sites. To better understand our results, we

have developed an original model in order to estimate the respective contributions to the net flux of the methanol adsorption/desorption in water films present in the ecosystem and of methanol degradation. We think this objective was met but we agree that it would be interesting in the future to link our model with a biogenic model to better quantify the contributions of the sources/sinks to the methanol exchanges.

P24012, line 16-20: Regarding air temperature, please give also numbers for October and spring 2010.

Yes, we will add into the manuscript these informations.

P24014, line 2: Due to the size of Fig. 2 it is hard to see the deposition during or following precipitation that is mentioned in the text. If the authors would like to highlight this specific observation an additional figure could be added to illustrate this link.

We will add points in figure 2 (see below) to identify easier when methanol deposition is accompanied with rains events.

P24019, lines 9-15: The authors in this study describe a very distinct uptake of methanol by the forest. One could expect that with a pattern this clear a similar sink should have been observed elsewhere at some point – even during considerably shorter measurement campaigns. It might be of interest to the authors that we also could not see a clear uptake of methanol over grassland at our study site in Neustift, Austria, in 2008 and 2009 (Hörtnagl et al., 2011), a site characterized by humid continental climate and plenty of dew formation.

Indeed, we were also very surprised. It is why in this study, we have developed a model to understand and to explain our measurements. We suppose that methanol depositions are probably also present at other sites but not in the same proportions. It would be the task of a site inter-comparison study to investigate these aspects.

Your long-term study is also very interesting but unfortunately, both studies were not focused on the same ecosystem type. Several hypotheses may be advanced to explain this difference. Firstly, vegetation of a grassland ecosystem is continuously growing in contrast with forest ecosystem, biogenic emissions from grassland may hide the methanol deposition on water surface. Secondly, to observe a significant deposition, you must have a destruction mechanism; it is possible that your site is not peopled with methylophilic bacteria for example.

We will change the sentence (P24005 line 26): "..., but none of them proposed a year-round follow-up of the exchange." by "..., but none of them (at the exception of Hörtnagl et al., 2011 above a temperate mountain grassland) proposed a year-round follow-up of the exchange.

P24038, Fig. 7: Please explain the negative values for M_{aw} (negative concentration values?).

The negative value is due to the modeled uncertainty of M_{aw} ($\pm 20 \text{ mg m}^{-3}$), we will add into the figure caption a short explanation.

Technical corrections

All technical corrections proposed by Hörtnagl have been applied.

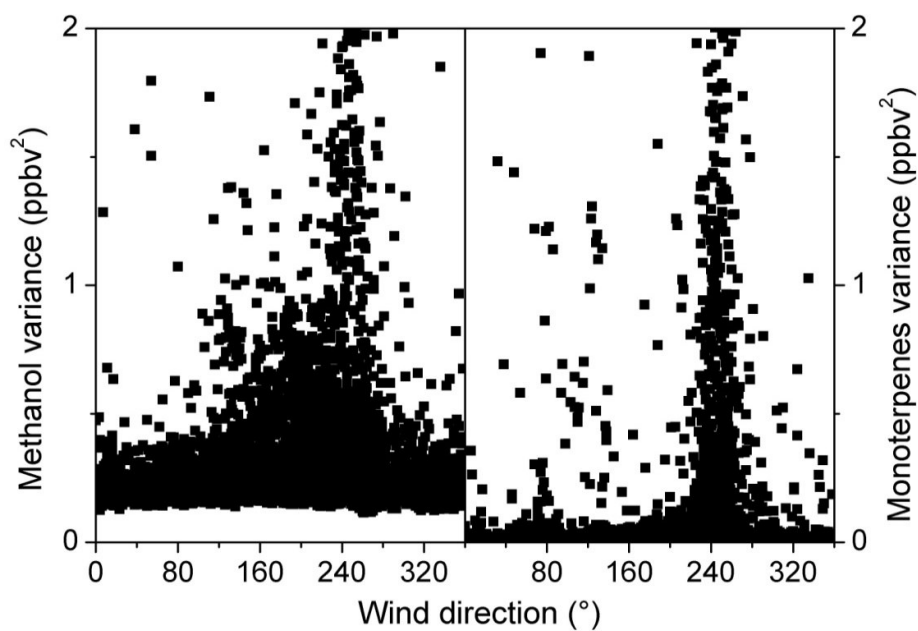


Figure A.

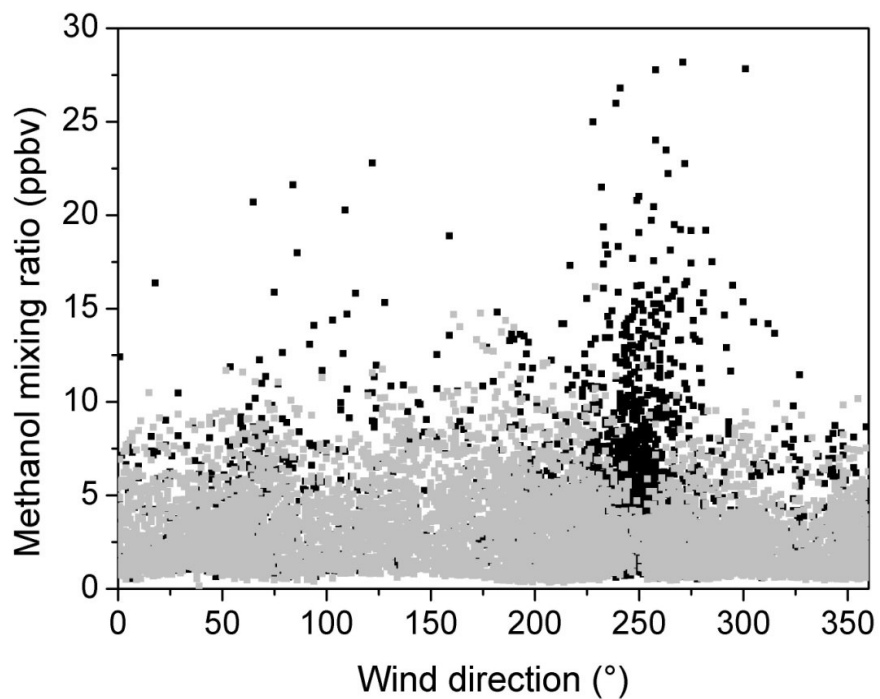


Figure B. The black points represent the data (2009-2010) deleted by the monoterpenes variance filtering. The light grey points represent the successful filtering data.

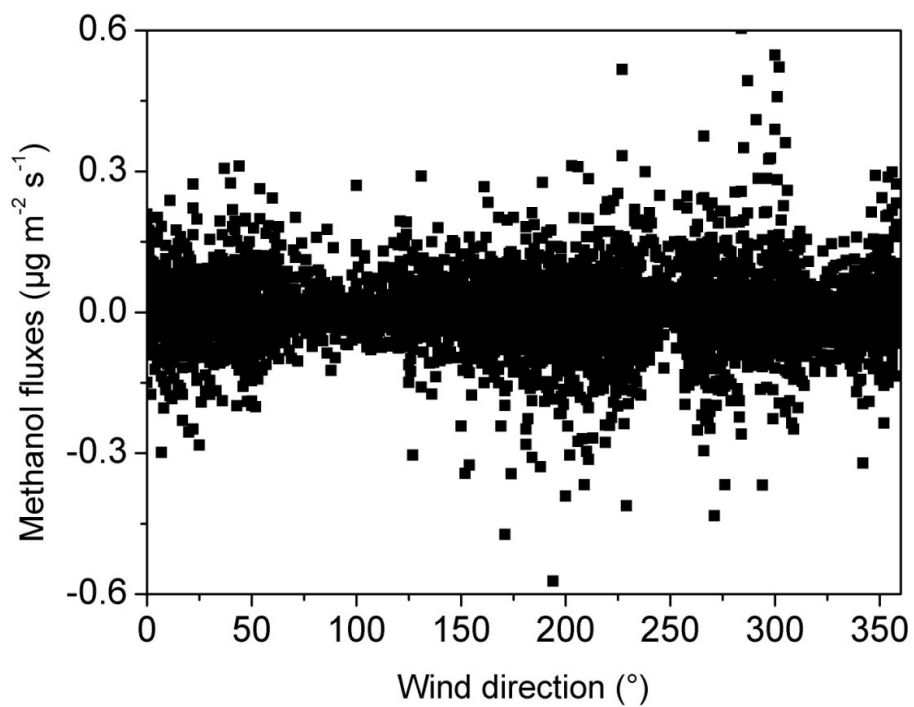


Figure C.

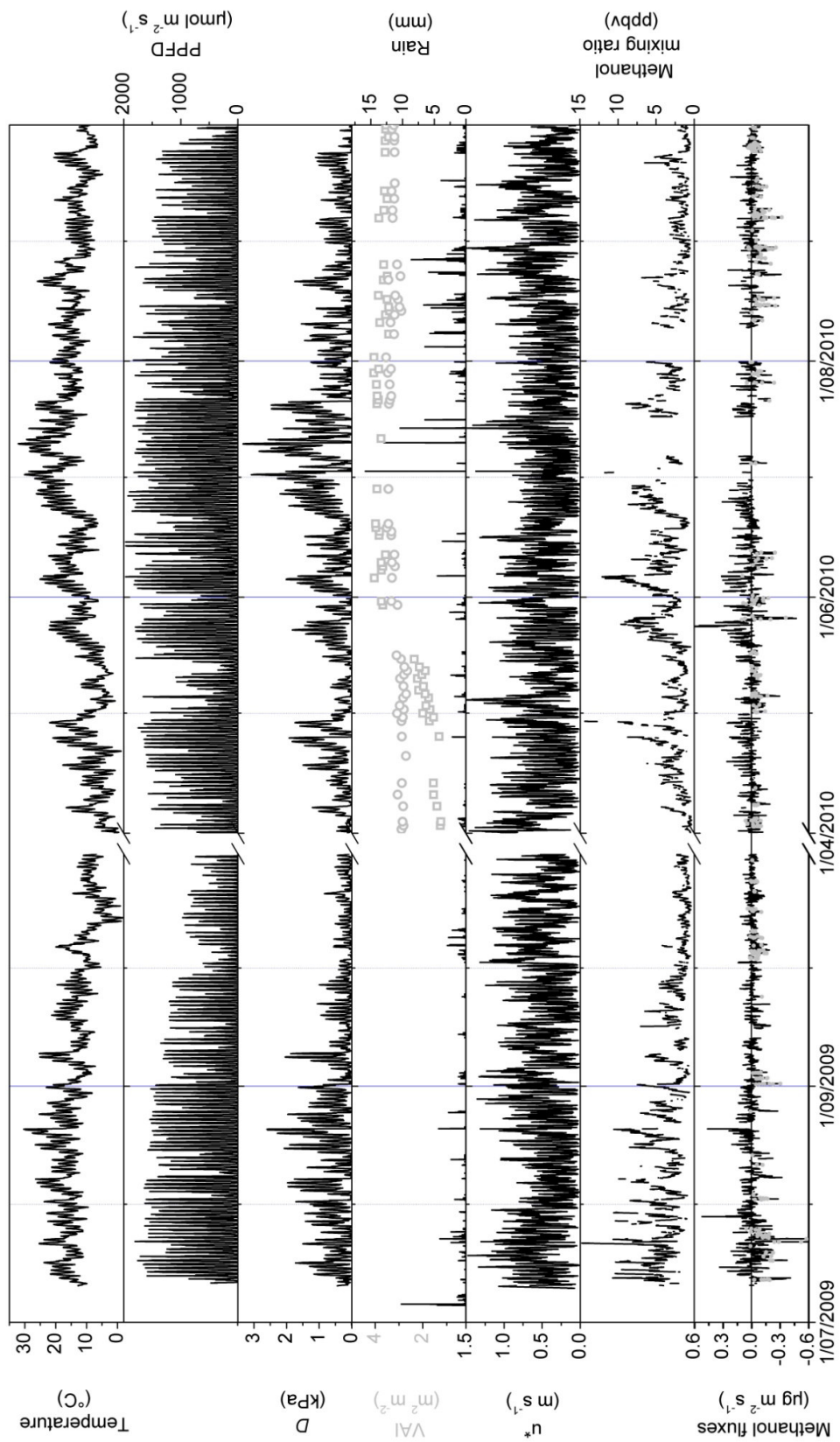


Figure 2.