We thank Dr. Holst for the constructive comment and the careful assessment of our paper. Please find below a point-by-point reply to your comments (italic: reviewer comments).

P24007, L3pp: as the forest stand is quite tall, you were sampling from the top of a 52m tower with a tubing length of 60m and an inner diameter of 6.4mm. This is a high surface to volume ratio for the sampling line, with a risk of wall losses especially for the methanol signal. Could you provide some more details about the sample line heating mentioned? Did you consider any kind of tests to see if there is significant influence of wall effects to methanol (i.e. in comparison to other VOCs presented in Laffineur et al. 2011)?

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."

A chromatographic effect for compounds having a high affinity to the tube surfaces can be checked with the time delay. In your study, the frequency distribution of the observed time delays for all BVOCs measured exhibited a peak near 14.8 s (this value was also confirmed by injection of isopropyl alcohol in the tube to determine the residence time). Spirig et al. (2005) has demonstrated that there were no significant differences (for methanol, isoprene and monoterpenes) in the attenuation of one VOC relative to another in a long inlet tube (similar to our tube characteristics but with a line heating at 5°C above the ambient temperature). We will add this explanation into the manuscript.

Chapter 2.2: can you provide some comments on calibration and determination of instrument background (zero air measurements)? This might be affecting the measured fluxes/concentrations directly and should therefore mentioned here instead of only referring to the Laffineur et al. 2011 paper.

We will add into the chapter 2.2 these sentences: "During the measurements, the instrumental background was determined every 4 h by sampling BVOC-free air, obtained by sending ambient air through a heated catalytic converter for 15 min (the last 8 min being used for the calculation of the mean background values). The background measurements for m/z 33 (protonated methanol) may be somewhat more complicated than the background measurements for the other compounds. Indeed, the measured background signal at m/z 33 consists of the real instrumental background at m/z 33 and the oxygen isotopes ($^{16}O^{17}O^+$) (Spirig et al. 2005). Background measurement was generated from ambient air just at the bottom of the tower, which can be somewhat more humid than the air from the top of the tower, which can have a small influence on the strength of the O_2^+ signal (m/z 32) and its second isotope. Once a month of 2010, we have estimated that the error caused by this effect on your m/z 33 measurements was less than 3%.
The sensitivity of the instrument was calibrated for the main target compounds (isoprene, sum of monoterpenes, methanol, acetone and acetaldehyde) every two or

three days using a gravimetrically prepared mixture of these gases in N_2 (Apel-Riemer Environmental, Denver, CO, USA) that contained approximately 500 ppbv isoprene, α-pinene and sabinene and about 1 ppmv methanol, acetaldehyde and acetone, with an accuracy of 5%. The compounds were further diluted (2-12 ppbv range) using a dynamic dilution system."

Chapter 2.4, wind direction: for the data filtering to exclude data possibly affected by anthropogenic influences you again refer to Laffineur et al. 2011, i.e. filtering was based on monoterpene concentration variance. Why not apply filtering based on the methanol concentration variance (assuming this effect shows for methanol as well) instead of using monoterpenes? You could then also analyse if filtering for monoterpenes and methanol exclude the same data points. At least, you should provide a figure like Fig. 2 from Laffineur et al. 2011 for methanol.

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.

Chapter 2.4, u*: you briefly discuss why you did not apply u* filtering here and refer to a publication which currently is not (yet) available, so I ask you to give some additional information here.

The book of Aubinet et al. 2011 will be available on January 2012 (Springer). Please find below an extract of Chapter 5 discussion about the night flux problems:
"…As the night flux problem results mainly from atmospheric processes that hinder the turbulent transport of tracers, it should affect any passive tracer that, similarly to CO_2, could be exchanged by the surfaces at night and whose flux would be mainly controlled by production /absorption mechanisms that carry out independently of the presence or absence of turbulent transport.

First, tracers whose fluxes are negligible at night, as water vapor and isoprene, could be considered as not concerned by such problem. For other tracers, like sensible heat, methane, monoterpenes, methanol, nitrous oxide, ozone or NOx the situation is more complex. In these cases, a careful and specific analysis is needed for each tracer to determine if the flux decrease under low turbulence (if any) is the result of a

measurement artifact or of a real flux slowing down. When the flux is not controlled by production / absorption processes at the surface but rather result from a diffusive exchange between a reservoir and the atmosphere, as is the case in deposition processes for example, the dependence of the flux on turbulence could be real. In these conditions, the night flux correction is not recommended for long term budgets as it could lead to a large flux overestimation…''

P24012, L15pp: if you mention the detailed mean temperatures and precipitation sums of the different periods involved, you should also give the numbers of the long-term averages to compare with!

Yes, you are right; we will add the annual mean temperatures and the annual cumulated precipitations for 2009 and 2010.

P24013, L14pp: changing wind directions (and possibly advection) in combination with a different species composition in the footprint area might have influenced deposition rates in the beginning and end of the night. Can you comment on this?

We need to develop a complementary study to understand exactly which processes reduce the deposition rates at the beginning of the night. At Vielsalm site, we know that the thermal stratification of the air is stronger in the beginning of the night than at the end. This stratification may increase the in-canopy aerodynamic resistance, may increase the footprint area and may produce advection. It is not the focus of this manuscript and the fluxes measured in the condition of strong thermal stratification is rejected from our analysis.
More generally, the deposition rates are probably influenced by the species that have not the same LAI and the same leaf/needle surface proprieties (influencing the water films formation). Footprint analysis did not permit to discern the impact of the species on the methanol depositions.

P24014, L2: you refer to Fig. 2 to prove that deposition is connected to periods with precipitation, but the scale of the figure is not really suitable to resolve this. Can you add a specific figure for this where this could be seen clearly?

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

P24014, L23: Could you add a figure for the relationship u* and methanol flux? Fig. 5 could be used here, but it provides flux/concentration against u*.

To avoid an overloading of the manuscript we prefer not to include another figure concerning methanol fluxes and u*. The relation between flux and u* will be not clear if you don't apply a selection criteria on the methanol concentration. The methanol concentration and u* drive together the deposition fluxes as shown by equation 1.

We will change our ambiguous sentence: '' The dependence of methanol deposition on u* indicate that…'' by ''The dependence of the deposition velocity on u* (Fig. 5) indicate that…''

P24016, L2: Would soil temperature measurements be available to get a better estimate of the water film temperature than air temperature (taken from which height a.g.l?)?

Yes, the soil temperature is available at Vielsalm but the temperature sensor closer to the soil surface is located at a depth of 10 cm that is not representative of the temperature at the soil surface level.

P24019, L9: You state that your model does not take LAI changes into account. This probably is most relevant during leaf unfolding and autumn, however, growth processes (and thus biogenic methanol emissions) take place during the whole growing season (without a significant change of LAI) and might thus be 'masked' by the adsorption/desorption and degradation processes in the model.

Exactly, there is a risk that biogenic emissions have been taken into account in the model calibration. This effect is probably limited because we have shown that the adsorption/desorption process of methanol in water films seems to be dominant on the biogenic emission in summer (calibration period).

Fig. 7: if Maw is the total methanol content in the water films, how can it be negative? To me, Fig. 7 seems to show the modelled uncertainty of Maw (which might be about +/- 20 mg/m3)?

Yes, it is. We will add to the figure caption this sentence: "The negative value is due to the modeled uncertainty of M_{aw} ($\pm$ 20 mg m^3)".

P24022, L14: Fig. 9 instead of Fig. 10?

Yes, you are right, we will modify that.

Fig. 1: could you briefly add explanations of variables shown to the caption?

Yes, we will add a description of variables.

Fig. 3: at least the summer 2010 figure is a mix of wet and dry periods. You show that deposition is mainly occurring at wet situations, so would it make sense to show an extra figure divided into wet and dry conditions?

I'm not sure that it makes sense; we know the air humidity conditions but not the soil surface humidity conditions. It is thus difficult to distinguish exactly wet and dry conditions. In figure 3, the wet and dry conditions are already partially divided because during the night the D values are generally lower than during the day.

Figs. 4-5, 10: I think A and B to mark panels are hardly visible.
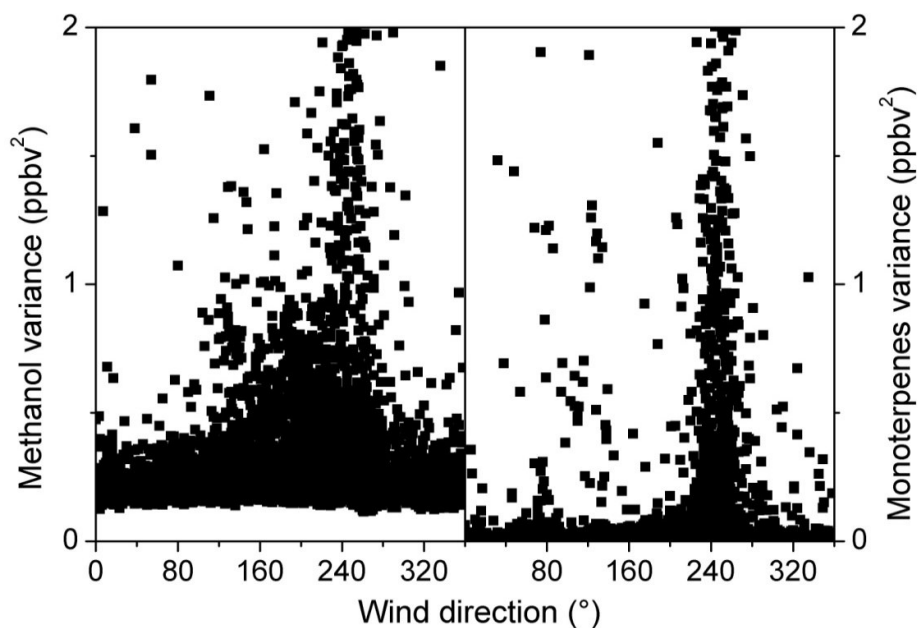
Yes, we will modify that.
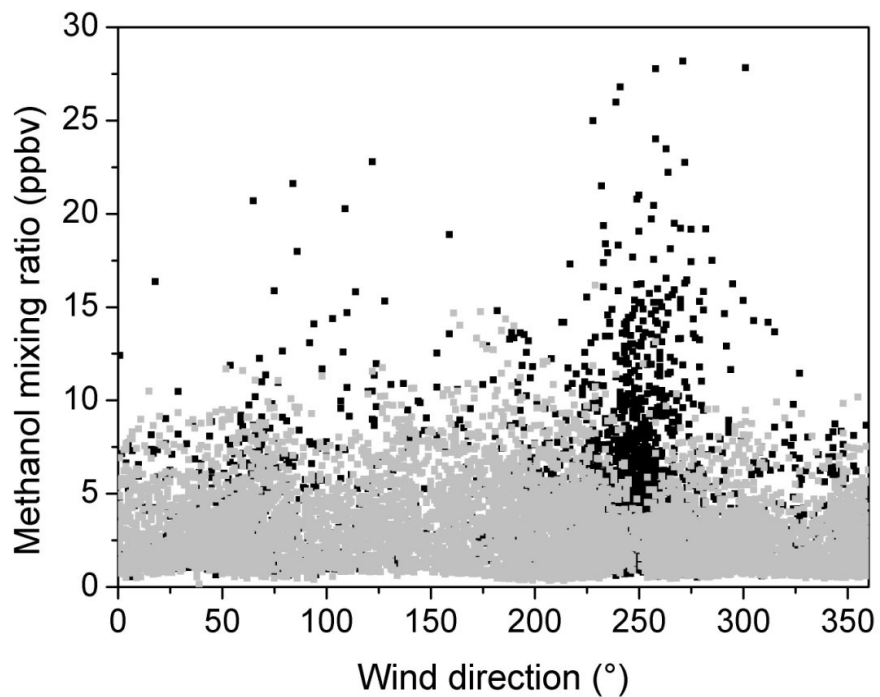

Figure A.

Figure B. The black points represent the data (2009-2010) deleted by the monoterpenes variance filtering. The light grey points represent the data points that succeed the filtering test.
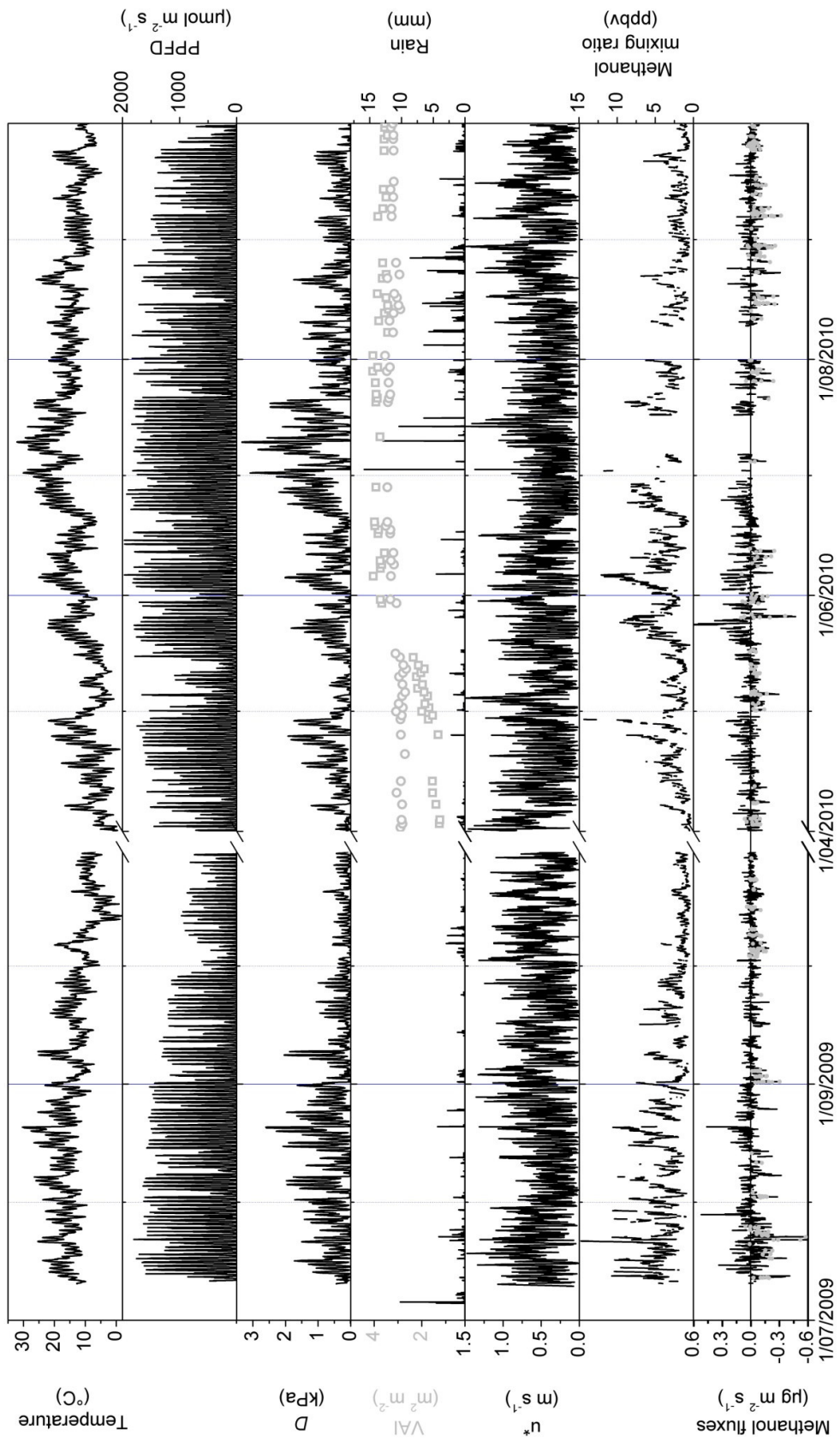
Figure 2.