## Reply to comments on "Boreal forest BVOCs exchange: emissions versus in-canopy sinks" by Anonymous Referee # 1

### October 21, 2017

We really appreciate the reviewer for the detailed and valuable comments which help us to improve the manuscript and extend the discussions in it. All the comments are replied below.

1. Comment: a) One of the main results of the study is the classification of selected BVOCs into different categories corresponding to the dominant source and sink terms. I recommend to add more discussion about the general validity of the classification of each compound, and to give more guidance about possible changes of this classification in different canopies.

Reply: Section 4.3 was split into two sections: "4.3 Overview of in-canopy sources and sinks" (from "The simulated monthly averaged ..." to "... given the low OH concentrations (Fig. 5c).") and "4.4 Classification of BVOCs" (from "Therefore the selected BVOCs ..." to "... the one proposed by Wesely (1989)."). More discussions about the general validity of the classification were added in the new section 4.4.

"The classifications of the featured BVOCs here can also be extended to other canopy types in summertime nearly without any modifications. For example, for isoprene+MBO, monoterpenes and sesquiterpenes the emission is always the only dominant local source within a canopy, although the emission potentials of these BVOCs can vary two or more orders of magnitude between different plant types (Guenther et al., 2012). Therefore, the current classifications for isoprene+MBO, monoterpenes and sesquiterpenes also apply to other canopy types.

Besides emission and dry deposition, acetaldehyde, methanol, acetone and formaldehyde can be chemically produced from the oxidation of other BVOCs and destroyed via OH oxidation or photolysis (Millet et al., 2010; Jacob et al., 2005; Khan et al., 2015; DiGangi et al., 2011). The chemical production and removal cancel out each other which can finally result in negligible net chemical effect as shown in this study (Fig. 5a). Therefore, the classifications of these four compounds also apply to other canopy types. However, further investigation with numerical simulations are still needed to verify the relative contributions of net chemical effects for different canopy types.

Chemical production is the only source in the planetary boundary layer for the other nonemitted gases, including acetol, pinic acid, BCSOZOH, ISOP34OOH, ISOP34NO3. They are either produced by direct chemical reactions inside the canopy or transported from above the canopy in all canopy types. Therefore, the classifications of them apply in a general way. "

2. b) I had difficulties following the discussion of many results because of the separation of turbulent transport and gas dry deposition, which is only briefly explained in the appendix. When used to parameterize canopy-scale flux measurements, dry deposition is typically a combination of these processes. Then, the deposition velocity is expressed as the inverse of the quasi-laminar sublayer resistance and the canopy resistance (rb and rc), AND the aerodynamic resistance (ra) derived from the turbulent diffusivity of the scalar of interest. The separation of turbulent transport and gas dry deposition should be clarified in the beginning of section 3.2, and the budget of sources and sinks should be introduced. Also, the definition of a deposition flux by Eq. 1 leads to some ambiguity throughout the manuscript. For example, in Figure 4 on p. 18 fluxes at the canopy top are discussed which are obviously not the deposition flux according to Eq. 1. Also, a downward turbulent flux (e.g. p. 20, line 20) is obviously defined as positive because it is a source with respect to the canopy. However, by micro-meteorological convention a downward turbulent flux (= deposition) would be negative. For clarity, these differences should be clearly pointed out throughout the manuscript.

Reply: 1. SOSAA has a multi-layer canopy module which is different from the big-leaf approach. The turbulent transport of scalars is explicitly computed by the model inside the canopy before they are deposited onto surfaces. Therefore, the aerodynamic resistance  $(r_a)$  is not explicitly included in our deposition scheme.

2. The introduction of source and sink terms was moved from appendix to the beginning of section 3.2.

3. The fluxes discussed in Fig. 4 are actually the upward turbulent fluxes at the canopy top, which are not the deposition fluxes. Now Eq. 1 was moved to the mass conservation equation and the definition of deposition flux was removed for clarity.

4. The concept used in this study is meant for a consistent discussion of sources and sinks of gases within the canopy, since the canopy is considered as a container.

P5, L25 - P7, L2:

" The gas dry deposition model is based on the O<sub>3</sub> dry deposition model described in Zhou et al. (2017). For each model layer, the deposition flux (F) of gas X is calculated as

$$F = -[X](\text{LAD}\Delta z V_{dveg} + A_s \Delta z V_{dsoil})$$
$$V_{dveg} = V_{dveg}(r_b, r_{stm}, r_{mes}, r_{cut}, r_{ws}, f_{wet})$$
$$V_{dsoil} = V_{dsoil}(r_{bs}, r_{soil})$$

where [X] is the concentration of gas species X,  $\Delta z$  is the layer thickness. LAD is the all-sided leaf area density at layer *i*.  $A_s$  represents the soil area index (Eq. 17 in Zhou et al., 2017).  $V_{dveg}$  is the vegetation layer-specific conductance which is a function of  $r_b$  (quasi-laminar boundary layer resistance),  $r_{stm}$  (stomatal resistance),  $r_{mes}$  (mesophyllic resistance),  $r_{cut}$  (dry cuticular resistance),  $r_{ws}$  (resistance to leaf wet skin) and  $f_{wet}$  (fraction of wet skin on leaf surface) (see Eqs. 8, 10 – 13 in Zhou et al. (2017)).  $V_{dsoil}$  is the soil conductance which is a function of  $r_{bs}$  (soil boundary layer resistance) and  $r_{soil}$  (soil resistance) (see Eq. 9 in Zhou et al. (2017)). "

 $\rightarrow$ 

<sup>&</sup>quot;The local change of the trace gas concentration at each model layer is determined by the gas emission  $(Q_{emis})$ , chemical production and loss  $(Q_{chem})$ , gas dry deposition  $(Q_{depo})$ , and turbulent transport flowing into or out of this layer  $(Q_{turb})$ . Here it should be noted that the positive (negative)  $Q_{turb}$  is a gas source (sink) term which indicates that the net effect of transportation increases (decreases) the gas concentration within the local layer. All of these processes are included in a mass conservation equation and are computed independently in the

model:

$$\frac{\partial[X]}{\partial t} = Q_{emis}^{t} + Q_{chem}^{t} + Q_{depo}^{t} + Q_{turb}^{t}$$

$$Q_{depo}^{t} = -[X](\text{LAD} \cdot V_{dveg} + A_{s}V_{dsoil})$$

$$Q_{turb}^{t} = \frac{\partial}{\partial z} \left( K \frac{\partial[X]}{\partial z} \right)$$

$$V_{dveg} = V_{dveg}(r_{b}, r_{stm}, r_{mes}, r_{cut}, r_{ws}, f_{wet})$$

$$V_{dsoil} = V_{dsoil}(r_{bs}, r_{soil})$$

Here  $Q_{emis}^t$  and  $Q_{chem}^t$  are directly calculated from the emission module and chemistry module in SOSAA, respectively. The superscript t represents instantaneous quantity. [X] (ng m<sup>-3</sup>) is the concentration of gas species X. LAD (m<sup>2</sup> m<sup>-3</sup>) is the all-sided leaf area density.  $A_s$ (m<sup>2</sup> m<sup>-3</sup>) represents the soil area index (Eq. 17 in Zhou et al., 2017). K (m<sup>2</sup> s<sup>-1</sup>) is the turbulent diffusivity for scalars.  $V_{dveg}$  (m s<sup>-1</sup>) is the vegetation layer-specific conductance which is a function of  $r_b$  (quasi-laminar boundary layer resistance; s m<sup>-1</sup>),  $r_{stm}$  (stomatal resistance; s m<sup>-1</sup>),  $r_{mes}$  (mesophyllic resistance; s m<sup>-1</sup>),  $r_{cut}$  (dry cuticular resistance; s m<sup>-1</sup>),  $r_{ws}$  (resistance to leaf wet skin; s m<sup>-1</sup>) and  $f_{wet}$  (fraction of wet skin on leaf surface; dimensionless) (see Eqs. 8, 10 – 13 in Zhou et al., 2017).  $V_{dsoil}$  is the soil conductance which is a function of  $r_{bs}$  (soil boundary layer resistance; s m<sup>-1</sup>) and  $r_{soil}$  (soil resistance; s m<sup>-1</sup>) (see Eq. 9 in Zhou et al., 2017). "

P29, L1: "Appendix A: Source versus sink terms"  $\rightarrow$  "Appendix A: Accumulated and integrated source and sink terms".

P29, L2–9: "The local change ... K is the turbulent diffusivity for scalars." was removed.

3. Comment: c) Figures 5-7 seem to indicate that the budget of in-canopy sources and sinks are balanced for almost all compounds for most of the time (when there is no white part in the bar charts). Then, according to Eq. A1 the in-canopy concentration tendency would be zero. Is this true?

Reply: Yes, it is true. All the source and sink terms are accumulated with time, e.g., emission and dry deposition. Therefore, the net in-canopy concentration tendency is neglected compared to these sources and sinks. For example, the in-canopy concentration change of methanol during the whole month is 2114.6  $\mu$ g m<sup>-3</sup> due to accumulated emissions, -3.5  $\mu$ g m<sup>-3</sup> due to chemical reactions, -406.5  $\mu$ g m<sup>-3</sup> due to turbulent transport, -1701.8  $\mu$ g m<sup>-3</sup> due to dry deposition. And the final concentration change is about 2.8  $\mu$ g m<sup>-3</sup> which is only 0.1% of the emission source.

## 4. Comment: d) On page 21, line 6 it is stated that emission sources shift upward during the day, "which implies that PAR and leaf temperature play a comparable role in emission rates besides the LAD". Why does the shift imply a comparable role?

Reply: Besides LAD, gas emissions are mainly dependent on leaf temperature and/or PAR, which are attenuated inside the canopy and thus decreasing gas emissions. If they play a minor role compared to LAD, the profiles of emission sources would be similar with the LAD profile. However, the peak heights of emission sources are about 50% higher than the LAD peak height, which indicates that PAR and leaf temperature also play an important role.

5. Comment: e) In my opinion, there is no additional benefit from the summary in Chapter 5. I recommend to revise this chapter, removing the purely summarizing parts and focusing on conclusions and synthesis.

Reply: The summary part was rewritten.

"Based on the  $O_3$  dry deposition model developed in Zhou et al. (2017), a new multi-layer gas dry deposition model extended from Wesely (1989) and Nguyen et al. (2015) was implemented into the 1D chemical transport model SOSAA. This model enabled the calculation of dry deposition processes within a forest canopy for all the gas compounds included in a chemistry scheme.

Using this model we analysed the monthly-averaged in-canopy sources and sinks of 12 featured BVOCs at SMEAR II in July, 2010. Several general behaviours of the selected BVOCs inside a boreal canopy were revealed. Throughout the whole day,  $\sim 86\%$  of monoterpenes and  $\sim 93\%$ of isoprene+MBO were transported out of the canopy after emitted from the canopy. However, canopy played as a sink for isoprene+MBO at nighttime when they were not emitted. On the contrary, most of the emitted sesquiterpenes were oxidized inside the canopy with only about 29% ventilated out. For the BVOCs with bi-directional fluxes, e.g., acetaldehyde, methanol, acetone and formaldehyde, a large portion or even all of the emitted gases were removed by dry deposition inside the canopy. The soil deposition contributed over 40% of the overall deposition sink. Moreover, the relative contributions of dry deposition sinks compared to emissions were maximum at dawn when highest RH values occurred. Acetol, pinic acid and BCSOZOH were removed by dry deposition inside the canopy with less than 20% compensated by chemical production, resulting in dominant downward fluxes throughout the whole day. ISOP34NO3 and ISOP3400H showed similar behaviours as acetol at nighttime. However, at daytime, the chemical productions from isoprene oxidation reactions were comparable with deposition sinks, which could even lead to a slightly upward flux at the canopy top at noon for ISOP34NO3.

The vertical distributions of in-canopy sources and sinks for all the gases had several common features. The vertical distribution of dry deposition onto vegetation surfaces always followed the LAD variation, which peaked at about 0.6  $h_c$  in this study. The peaks of emission sources were at 0.8 to 0.9  $h_c$  which was higher than dry deposition because the attenuated PAR and leaf temperature reduced the emission rate inside the canopy. The vertical profile of chemical sinks for sesquiterpenes were nearly the same as their emissions since they were mostly oxidized right after being emitted. The chemical productions for ISOP34NO3 and ISOP34OOH were maximum around the canopy top where the isoprene emission peaked and the radiation was high.

According to the significance of different source and sink terms, the selected BVOCs were classified into five categories: Cemis (monoterpenes, isoprene+MBO), Cemis-chem (sesquiterpenes), Cemis-depo (acetaldehyde, methanol, acetone, formaldehyde), Cdepo (acetol, pinic acid, BCSOZOH), Cchem-depo (ISOP34OOH, ISOP34NO3), where the subscripts represented the significant terms. Generally speaking, the classifications for these BVOCs were expected to be applicable in other canopy types at least in summertime.

This study has provided a method to quantify the proportion of dry deposition sinks for various BVOCs which can be applied in large-scale models in future. On the basis of the analysis of 12 selected BVOCs or groups of BVOCs in this study, a large amount of other compounds with similar properties can be represented by being classified into the five categories mentioned above. For example, OVOCs most likely belong to categories Cdepo and Cchem-depo, which indicates that dry deposition can not be neglected when their sources and sinks are investigated. In addition, the categories Cemis-chem and Cemis-depo imply that the simulation of individual processes is necessary to help further analyse the measured emission data of such gases, and thus obtain a more accurate estimation of BVOCs exchange.

This study has shown that dry deposition of oxidation products of precursor gases as well as other BVOCs could be a potentially important feature of improving our understanding and quantification of BVOCs exchange. However, such assessments are largely limited by available observations that could further corroborate the correctness of the simulated deposition processes as presented in this study. Furthermore, this study stresses the necessity of applying a canopy exchange modeling system for a detailed analysis of BVOCs exchange regimes within and above a boreal forest canopy, instead of applying a big-leaf representation without considering the interactions between emissions, chemistry, turbulent transport and deposition.

6. Comment: p.1, line 15: "Most of the simulated sources and sinks were located...": Without knowing the forest geometry, e.g. the canopy height, absolute heights are not very helpful. I recommend using the height above ground level relative to the canopy height, height/hc (similar to the presentation of Figure 2a and b) throughout the manuscript, and in particular also in Figures 8 and 9 and their discussions.

Reply:

P1, L15: "about 4 m"  $\rightarrow$  "about 0.2  $h_c$  (canopy height)".

- P1, L15: "about 8 m"  $\rightarrow$  "about 0.4  $h_c$ ".
- P1, L17: "about 14 16 m"  $\rightarrow$  "about 0.8 0.9  $h_c$ ".
- P1, L17: "than 10 m"  $\rightarrow$  "than 0.6  $h_c$ ".

P21, L22: "about 8 m"  $\rightarrow$  "about 0.4  $h_c$ ".

P21, L23: "about 4 m"  $\rightarrow$  "about 0.2  $h_c$ ".

P21, L24: "below 8 or 4 m"  $\rightarrow$  "below 0.4 or 0.2  $h_c$ ".

P23, L19-23: The whole summary section was rewritten, so the height levels were not converted to canopy height here.

Figures 8 and 9: The y axes were changed to "Height/ $h_c$ ". Several x scales were modified to zoom in the plots in x direction (figures are placed at last).

### 7. Comment: p.1, line 21: "in-canopy" instead of "in-caonpy"

Reply: P1, L21: "in-caopy"  $\rightarrow$  "in-caopy".

## 8. Comment: p.1, lines 22/23: I recommend to divide the last sentence of page 1 into two sentences.

Reply: P1, L22–23: "Twelve featured BVOCs or BVOC groups were analyzed in this study, more compounds could also be investigated similarly by being classified into the five categories."

 $\rightarrow$ 

"Twelve featured BVOCs or BVOC groups were analyzed in this study. Other compounds could also be investigated similarly by being classified into these five categories.".

#### 9. Comment: p.2, line 16: A comma is missing after methanol.

Reply: P2, L16: "methanol acetaldehyde"  $\rightarrow$  "methanol, acetaldehyde".

- 10. Comment: p.2, line 19: Revise "... or partly transported into higher atmosphere". Reply: P2, L19: ", or partly transported into higher atmosphere."  $\rightarrow$  ", or transported throughout the planetary boundary layer".
- 11. Comment: p.2, line 24: "leaf-scale" instead of "leaf-sale" Reply: P2, L24: "leaf-sale"  $\rightarrow$  "leaf-scale".
- 12. Comment: p.2, line 29: You may also add a reference to Bamberger et al. (2011) Deposition fluxes of terpenes over grassland. J. Geophys. Res. 116, D14305, doi:10.1029/2010JD015457.

Reply: P2, L30: "... in field measurements."

 $\rightarrow$ 

"... in field measurements. Bamberger et al. (2011) observed the deposition fluxes of monoterpenes, sesquiterpenes and oxygenated terpenes over a temperate mountain grassland in an alpine valley after a hailstorm."

## 13. Comment: p.3, line 10: Could you please add a reference for the GECKO-A model?

Replay: A reference was added and a typo was also corrected in the same sentence.

P3, L10–12: "The models GECKO-A (Generator of Explicit Chemistry and Kinetics of Organics in the Atmosphere) and GROMHE (Raventos-Duran et al., 2010, GROup contribution Method for Henry's law Estimate;) ..."

 $\rightarrow$ 

"The models GECKO-A (Generator of Explicit Chemistry and Kinetics of Organics in the Atmosphere; Aumont et al., 2005) and GROMHE (GROup contribution Method for Henry's law Estimate; Raventos-Duran et al., 2010) ..."

## 14. Comment: p.3, lines 22/23: What do you mean by: "Both of them motivated this study"?

Reply: As we mentioned in the same paragraph: the models, which calculated the deposition sinks of a large number of BVOCs, were big-leaf models. The other models, which were multi-layer canopy models, only considered a small amount of BVOCs (usually less than 100 gas species). So these gaps motivated this study. This sentence here was not clear, so we deleted it and modified the first sentence in next paragraph.

P3, L22-23: "Moreover, detailed deposition contributions for BVOCs have not been analysed. Both of them motivated this study."

 $\rightarrow$ 

"Moreover, detailed deposition contributions for BVOCs have not been analysed."

P3, L24: "A multi-layer gas dry deposition model has been developed in this study based on several models in previous studies ..."

 $\rightarrow$ 

"In order to fill the gaps mentioned above, a multi-layer gas dry deposition model has been developed in this study based on several models in previous studies ..."

### 15. Comment: p.3, line 29: "complementary" instead of "complementory"

Reply: P3, L29: "complementary"  $\rightarrow$  "complementary"

# 16. Comment: p.4. line 29: I cannot follow the explanation of the data filtering: A one month-data set of 45-minute averages would consist of almost 1000 data points for each time series. Why did you filter out one data point "from 164 measurement data points"?

Reply: In 2010, BVOC concentrations were measured by PTR-MS at six levels (4.2 m, 8.4 m, 16.8 m, 33.6 m, 50.4 m and 67.2 m) one by one in one single measurement cycle (6 minutes). Then nine cycles including one cycle for determining instrumental background were conducted every third hour. The 45-minute averages of BVOC concentrations were computed from nine cycles. Finally, the BVOC fluxes were calculated from the 45-minute average data with the surface-layer-profile method (Rantala et al., 2014, 2015). So the flux data were computed every third hour, containing at most 248 points in July, 2010. 164 measurement data were available after excluding the periods when instruments worked improperly. This sentence was modified to be more clear.

P4, L29: "The fluxes of BVOCs, based on 45-minute averages, were computed with the surface-layer-profile method ..."

 $\rightarrow$ 

"The fluxes of BVOCs, based on 45-minute averages of BVOC concentrations, were computed every third hour with the surface-layer-profile method ...".

17. Comment: p.6, Table 1: In the first and second columns, "delta3-pinene" should read "delta3-carene"

Reply: P6, Table 1: " $\Delta^3$ -pinene"  $\rightarrow$  " $\Delta^3$ -carene".

- 18. Comment: p.11, line 31: Remove "." after "BVOCs" Reply: P11, L31: "BVOCs. (Table ..."  $\rightarrow$  "BVOCs (Table ..."
- 19. Comment: p.12, Table 3: In the first column, "delta3-pinene" should read "delta3carene"

Reply: P12, Table 3: " $\Delta^3$ -pinene"  $\rightarrow$  " $\Delta^3$ -carene".

20. Comment: p.13, Table 4: In the first column, "delta3-pinene" should read "delta3carene"

Reply: P13, Table 4: " $\Delta^3$ -pinene"  $\rightarrow$  " $\Delta^3$ -carene".

21. Comment: p.14, line 10: The statement that in "the sub-canopy (0.6 m) the incoming PAR was only about 1/4 of that at the canopy top" is not evident from Figure 1b. Is this statement with respect to maximum values or daily sums or something else? Just by looking at Figure 1b, most subcanopy values seem to be around 1/3.

Reply: It describes the monthly average PAR values. The text was modified.

P14, L7: "the incoming PAR was only about 1/4"  $\rightarrow$  "the monthly-averaged incoming PAR was only 1/4".

22. Comment: p.14, line 9: Is the accuracy of the precipitation measurement good enough to give a value of 34.64 mm for the accumulated precipitation?

Reply: The measured 1-minute accumulated precipitation data from SMEAR II data server (downloaded from https://avaa.tdata.fi/web/smart/smear/search) has the precision of 0.01 mm. However, according to the manual of the instrument Vaisala FD12P

(https://www.manualslib.com/manual/538824/Vaisala-Fd12p.html), the accuracy of precipitation intensity is  $\pm 30\%$  in the range of 0.5 to 20 mm h<sup>-1</sup>. So the accuracy of accumulated precipitation during previous 1 minute is in the range of 0.008 to 0.3 mm. We modified the number with less precision.

P14, L9: "34.64 mm"  $\rightarrow$  "35 mm".

- 23. Comment: p.15, line 8: "due to the buoyancy" instead of "due to buoancy" Reply: P15, L8: "due to buoancy"  $\rightarrow$  "due to the buoyancy".
- 24. Comment: p.15, line 11: "occurrence" instead of "occuring" Reply: P15, L11: "occuring"  $\rightarrow$  "occurrence".
- 25. Comment: p.15, line 16: Is this reference to Section 4.4 pointing to line 26 on page 20? I couldn't find a true demonstration of the potentially important role of leaf wetness in Section 4.4. Also, it should be acknowledged that leaf wetness may play a role at RH < 70 %, e.g. depending on the deliquescent behavior of salts deposited on the vegetation surfaces.

Reply: We removed the latter part of the sentence and added the impact of deliquescence in the end of the paragraph.

P15, L14-16: "Therefore, the observed RH values inside the canopy were used to parametrise  $f_{wet}$  when calculating the deposition velocity to represent a more realistic leaf wetness condition, also since this leaf wetness plays a potentially important role in BVOC exchange as we demonstrate in further details below in Section 4.4."

 $\rightarrow$ 

"Therefore, the observed RH values inside the canopy were used to parametrise  $f_{wet}$  when calculating the deposition velocity to represent a more realistic leaf wetness condition. It should be noted here that although RH = 70% is chosen as a threshold of the occurrence of leaf wetness in the model, the leaf wetness may already play a role when RH < 70%, e.g., due to the deliquescent effect of deposited salt on the vegetation surfaces."

#### 26. Comment: p.16, line 8: Remove "from the ecosystem".

Reply: P16, L8: "from the ecosystem into the soil"  $\rightarrow$  "into the soil".

27. Comment: p.17, line 5: I would assume that the standard deviation of the measured data shown in Figure 4b is not only due to measurement uncertainties but also due to day-to-day variability over the course of the month. If this is the case, please rephrase "measurement uncertainties".

Reply: P17, L5: "measurement uncertainties"  $\rightarrow$  "measurement uncertainties and day-to-day variation".

28. Comment: p.19, line 4: Revise "still keep similar".

Reply: P19, L4: "... emitted compounds still keep similar except for ..."  $\rightarrow$  "... emitted compounds are similar with that at daytime except for ...".

- 29. Comment: p.19, line 17: "In the second..." instead of "While in the second..." Reply: P19, L17: "While in the second category, ..."  $\rightarrow$  "In the second category, ...".
- 30. Comment: p.21, line 9: "... inside the canopy compared to the vertical..." instead of "... inside the canopy compared to the effect of vertical..."
  Reply: P21, L9: "... inside the canopy compared to the effect of vertical temperature gradient ..." → "... inside the canopy compared to the vertical temperature gradient ...".
- 31. Comment: p.21, line 21: "most" instead of "a majority portion of" Reply: P21, L21: "a majority portion of"  $\rightarrow$  "most".
- 32. Comment: p.22, line 1: "chemical sources" instead of "chemical productions" Reply: P22, L1: "chemical productions"  $\rightarrow$  "chemical sources".
- 33. Comment: p.22, line 4: "phenomenon" instead of "phenomena" Reply: "phenomena"  $\rightarrow$  "phenomenon".
- 34. Comment: p.23, lines 5/6: Revise "...need of the application of canopy exchange modelling system..."

Reply: The whole summary section was rewritten (see Comment 5)

35. Comment: p.23, line 15: "causing a slight upward flux" instead of "causing slightly upward flux"

Reply: The whole summary section was rewritten (see Comment 5)

36. Comment: p.24, lines 2-5: This sentence basically repeats p.23, lines 5-7. Reply: The whole summary section was rewritten (see Comment 5)

## References

- Aumont, B., Szopa, S., and Madronich, S. (2005). Modelling the evolution of organic carbon during its gas-phase tropospheric oxidation: development of an explicit model based on a self generating approach. Atmospheric Chemistry and Physics, 5(9):2497–2517.
- Bamberger, I., Hörtnagl, L., Ruuskanen, T. M., Schnitzhofer, R., Müller, M., Graus, M., Karl, T., Wohlfahrt, G., and Hansel, A. (2011). Deposition fluxes of terpenes over grassland. *Journal of Geophysical Research: Atmospheres*, 116(D14). D14305.
- DiGangi, J. P., Boyle, E. S., Karl, T., Harley, P., Turnipseed, A., Kim, S., Cantrell, C., Maudlin III, R. L., Zheng, W., Flocke, F., Hall, S. R., Ullmann, K., Nakashima, Y., Paul, J. B., Wolfe, G. M., Desai, A. R., Kajii, Y., Guenther, A., and Keutsch, F. N. (2011). First direct measurements of formaldehyde flux via eddy covariance: implications for missing in-canopy formaldehyde sources. *Atmospheric Chemistry and Physics*, 11(20):10565–10578.
- Guenther, A. B., Jiang, X., Heald, C. L., Sakulyanontvittaya, T., Duhl, T., Emmons, L. K., and Wang, X. (2012). The Model of Emissions of Gases and Aerosols from Nature version 2.1 (MEGAN2.1): an extended and updated framework for modeling biogenic emissions. *Geosci. Model Dev.*, 5:1471–1492.
- Jacob, D. J., Field, B. D., Li, Q., Blake, D. R., de Gouw, J., Warneke, C., Hansel, A., Wisthaler, A., Singh, H. B., and Guenther, A. (2005). Global budget of methanol: Constraints from atmospheric observations. Journal of Geophysical Research: Atmospheres, 110(D8):n/a-n/a. D08303.
- Khan, M., Cooke, M., Utembe, S., Archibald, A., Maxwell, P., Morris, W., Xiao, P., Derwent, R., Jenkin, M., Percival, C., Walsh, R., Young, T., Simmonds, P., Nickless, G., O'Doherty, S., and Shallcross, D. (2015). A study of global atmospheric budget and distribution of acetone using global atmospheric model STOCHEM-CRI. *Atmospheric Environment*, 112(Supplement C):269– 277.
- Millet, D. B., Guenther, A., Siegel, D. A., Nelson, N. B., Singh, H. B., de Gouw, J. A., Warneke, C., Williams, J., Eerdekens, G., Sinha, V., Karl, T., Flocke, F., Apel, E., Riemer, D. D., Palmer, P. I., and Barkley, M. (2010). Global atmospheric budget of acetaldehyde: 3-D model analysis and constraints from in-situ and satellite observations. *Atmospheric Chemistry and Physics*, 10(7):3405–3425.
- Nguyen, T. B., Crounse, J. D., Teng, A. P., St. Clair, J. M., Paulot, F., Wolfe, G. M., and Wennberg, P. O. (2015). Rapid deposition of oxidized biogenic compounds to a temperate forest. *PNAS*, 112(5):E392–E401.
- Rantala, P., Aalto, J., Taipale, R., Ruuskanen, T. M., and Rinne, J. (2015). Annual cycle of volatile organic compound exchange between a boreal pine forest and the atmosphere. *Biogeosciences*, 12(19):5753–5770.
- Rantala, P., Taipale, R., Aalto, J., Kajos, M. K., Patokoski, J., Ruuskanen, T. M., and Rinne, J. (2014). Continuous flux measurements of VOCs using PTR-MS - reliability and feasibility of disjunct-eddy-covariance, surface-layer-gradient, and surface-layer-profile methods. *Boreal Envi*ronment Research, 19:87–107.
- Raventos-Duran, T., Camredon, M., Valorso, R., Mouchel-Vallon, C., and Aumont, B. (2010). Structure-activity relationships to estimate the effective henry's law constants of organics of atmospheric interest. Atmospheric Chemistry and Physics, 10(16):7643–7654.
- Wesely, M. L. (1989). Parameterization of surface resistances to gaseous dry deposition in regionalscale numerical models. Atmos. Env., 23:1293–1304.

Zhou, P., Ganzeveld, L., Rannik, Ü., Zhou, L., Gierens, R., Taipale, D., Mammarella, I., and Boy, M. (2017). Simulating ozone dry deposition at a boreal forest with a multi-layer canopy deposition model. Atmospheric Chemistry and Physics, 17:1361–1379.



Figure 8: Modeled monthly-averaged vertical profiles of weighted  $\overline{Q}_n^{\Delta}$  (solid lines) and the relative contributions  $\overline{Q}_{rel,n}^{\Delta}$  (bars) of gas emissions (green), net chemical production and loss (red), turbulent transport (blue) and gas dry deposition (purple) within the canopy for (a) monoterpenes, (b) isoprene+MBO, (c) acetaldehyde, (d) sesquiterpenes, (e) methanol and (f) acetone. The second panels (g) to (l) and the third panels (m) to (r) are for the same compounds but the average is done for daytime and nighttime, respectively. The height is normalised by the canopy height  $(h_c)$ . The values of weighted  $\overline{Q}_n^{\Delta}$  at surface layer are divided by 10 for clarity. The original values at surface layer for deposition (left) and transport (right) are shown as float numbers at the bottom for each plot. The x labels and y labels of the left bottom subfigure also apply to all the other subfigures.



Figure 9: The same as Fig. 8 except for formaldehyde, acetol, pinic acid, BCSOZOH, ISOP34NO3 and ISOP34OOH. Note that the bottom numbers for BCSOZOH, ISOP34NO3 and ISOP34OOH also needs to be scaled by  $10^{-5}$ ,  $10^{-5}$  and  $10^{-4}$ , respectively.