Responses to the comments of Referee#2

General comments:

This paper presents the atmospheric loadings and seasonal distributions of secondary fatty alcohols in size-resolved aerosols from a deciduous forest canopy at Hokkaido, Japan. The results obtained have been interpreted logically. Based on the distributions of n-nonacosan-10-ol and its non-linear relations with the primary organic aerosol markers in the same samples such as sucrose, trehalose, mannitol and arabitol in spring, summer and autumn together with the comparisons of the SFAs measured in plant leaves, authors have found that the SFAs are derived from plant waxes. Overall, the data presented here and the conclusions drawn from them are interesting and make a substantial contribution to the community of atmospheric- and biogeo-sciences. Therefore, I recommend this paper for final publication in BG, after addressing the following comments.

Reply: We appreciate the referee's valuable comments on our work. Our responses to the specific comments and details of the changes made to the manuscript are given below.

Specific comments:

1. Section 3.1 - Page 4, Lines 12-15: The average concentrations of SFA3 --- are smaller than those --- (Miyazaki et al., 2019). Here it is important to note the literature values and describe the potential reasons behind such lower levels (10~20 times) compared to those observed at Tomakomai experimental forest.

Reply 1: In total suspended particulate (TSP) matter obtained previously at Tomakomai (TMK) experimental forest (42°43′ N, 141°36′ E), the average concentrations of *n*-nonacosan-10-ol (SFA3) and *n*-nonacosan-5,10-diol (SFA5) were ~100 ng m^{-3} and ~8 ng m^{-3}, respectively, in spring (Miyazaki et al., 2019). These concentrations are 10–15 times larger than those at this study site (42°59′ N, 141°23′ E). The difference in the concentration levels is likely due to several factors: the area of TMK experimental forest (2715 ha) is substantially larger than that (147 ha) of the forest site in this study. Moreover, leaf area index (LAI) of TMK

(~4–6) (Hiura, 2001) is larger than that of this study site (<4) (Nakai et al., 2003). These suggest that the TMK experimental forest has more biomass, which has a potential to emit SFAs, than that in the current study site. Furthermore, TMK consists of a wide variety of vegetation including mature and secondary deciduous trees and man-made coniferous trees, the number of which is much larger than that of this study site. This implies that TMK has much more potential to emit PBAPs, although dominant vegetation species that emit SFAs has not been specifically identified. Taking account of the referee's comment, we made additional statement with the values in literature as follows:

Page 4, Lines 12: ***"The average concentrations of SFA3 (10.2±10.3 ng m^{-3}) and SFA5 (0.52±0.69 ng m^{-3}) in the bulk aerosol (the sum of submicrometre and supermicrometre aerosol mass) during spring are smaller than those (~100 ng m^{-3} and ~8 ng m^{-3}, respectively) in total suspended particulate (TSP) matter obtained at Tomakomai (TMK) experimental forest (42°43′ N, 141°36′ E) (Miyazaki et al., 2019). If most of these SFAs were originated from forest vegetation, the difference in the concentration levels is attributable to the larger area (2715 ha) and leaf area index (LAI; ~4–6) with much more plant species in the TMK experimental forest (Hiura, 2001) compared to those (150 ha and <4, respectively) of the current research forest site (Nakai et al., 2003). "***

2. Section 3.3.1 – Page 8, Lines 13-14: Correlations between --- two seasons (Fig. 6a). It is contrary to a significant correlation (R2 = 0.70) reported between the *n*-nonacosan-10-ol and sucrose in forest aerosols by Miyazaki et al., 2019. So, I suggest the authors to compare the data (the sum of submicrometre and supermicrometre aerosol mass) with that (in TSP) reported by Miyazaki et al., 2019 in order to make it clear.

Reply 2: Although the correlation coefficient between *n*-nonacosan-10-ol and sucrose was discussed in both studies, time scales and size bins of data used for the calculation in the two studies are completely different. The significant correlation shown in Miyazaki et al. (2019) was based on the time series data of TSP on a time scale of one year. The correlation shown in their study meant that the seasonal trends of the SFA and sucrose concentrations in TSP were similar, both of which showed peaks in the growing season. On the other hand, the relation between *n*-nonacosan-10-ol and sucrose in our study was based on the data of each size bins obtained in one week of each season (spring or summer). The insignificant

correlation meant that the size distributions with peaks were different between the two compounds, suggesting the different sources. Although it is difficult to calculate correlation coefficient for TSP in this study because of the limited number of data (three in one season), the seasonal trend of the *n*-nonacosan-10-ol and sucrose concentrations in TSP in our study were similar to that reported in Miyazaki et al. (2019) (i.e., maximum in spring).

References:

Hiura, T.: Stochasticity of species assemblage of canopy trees and understory plants in a temperate secondary forest created by major disturbances, *Ecol. Res.*, 16, 887–893, 2001.

Miyazaki, Y., Gowda, D., Tachibana, E., Takahashi, Y., and Hiura, T.: Identification of secondary fatty alcohols in atmospheric aerosols in temperate forests, *Biogeosciences*, 16, 2181-2188, https://doi.org/10.5194/bg-16-2181-2019, 2019.

Nakai, Y., Kitamura, K., Suzuki, S., and Abe, S.:Year-long carbon dioxide exchange above a broadleaf deciduous forest in Sapporo, Northern Japan, *Tellus*, 55B, 305–312, 2003.