

We appreciate the referee's valuable comments on our work. Our responses to the specific comments are given below.

Responses to the comments of Referee#1:

Comment 1: Page 30778 – line 12: a reference to a previous study of the authors, in which the analysis by ion chromatography is reported in sufficient detail, is lacking here.

***Reply 1:* A reference of an ion chromatography analysis (Miyazaki et al., 2009) is now given in the text.**

Comment 2: Page 30779 – line 8: Not all readers interested in this article will be acquainted with the term “C3 plant origin”. I suggest to briefly explain this term.

***Reply 2:* We have added an explanation about C3 plants and their $\delta^{13}\text{C}$ to the revised manuscript as suggested:**

“For example, C3 plants, which use the Calvin-Benson cycle as a metabolic pathway for carbon fixation in photosynthesis, have $\delta^{13}\text{C}$ values typically in the range of -23 to -30‰ (Gelencsér, 2004). All trees and most shrubs, grasses, and sedges in mid-latitude and boreal regions belong to the C3 class of plants.”

Comment 3: Page 30780 – lines 12-14: It would be appropriate to provide some references for the biogenic SOA markers; some of them are already rather well known but others like MBTCA are less well established:

- 2-methylerythritol and 2-methylthreitol (Claeys et al., 2004)
- pinic acid (e.g., Yu et al., 1999)
- MBTCA (Szmigielski et al., 2007). The latter article is referred to later in the text but it is relevant to already mention it at this early stage
- 3-HGA (Claeys et al., 2007)

***Reply 3:* All the references for the biogenic SOA tracers given by the referee have been added to the text as suggested.**

Comment 4: Page 30781 – line 12: 3-HGA showed the highest concentrations throughout the study period. It would be relevant to compare this result with other field studies where 3-HGA was measured. For example, 3-HGA also showed the highest concentrations (in ng/m^3 ; median, 16.8; average, 19.7; range: 5.4–113) among the α -/ β -pinene SOA tracers for $\text{PM}_{2.5}$ aerosols that were collected during a 2003 spring-summer campaign in K-puszt,

Hungary (Kourtchev et al., 2009).

Reply 4: As suggested, we have included a description of the results of Kourtchev et al. (2009) for comparison: “Kourtchev et al. (2009) reported that 3-HGA also showed the highest concentrations (with a median value of 16.8 ng m^{-3}) among the α -/ β -pinene SOA tracers for $\text{PM}_{2.5}$ aerosols collected at a mixed coniferous/deciduous forest site in K-puszta, Hungary, during summer 2003.”

Comment 5: Page 30781 – lines 19-25: Trehalose is together with the sugar alcohols, arabitol and mannitol, a known constituent of fungal spores and fragments (Bielecki, 1982; Lewis and Smith, 1967). I therefore suggest that a more clear link is made here to fungal spores and fragments as a possible primary source. The authors mention resuspended soil and associated biota, which is rather vague. If the authors have arabitol and mannitol concentrations, it would also be very relevant to compare them with those of trehalose and to see whether there is a correlation. In this way, they could provide additional support that the primary source of trehalose is indeed fungal spores and fragments.

Reply 5: We appreciate the referee’s helpful comment. Indeed, we measured the concentrations of arabitol and mannitol, both of which showed positive correlations with those of trehalose. We fully agree that the result provides additional support for the idea that fungal spores and fragments are the primary source of trehalose. This has been clearly mentioned in the revised manuscript:

“In fact, the trehalose concentration showed positive correlations with the concentrations of arabitol ($r^2 = 0.55$) and mannitol ($r^2 = 0.77$), which are major fungal polyols in many green algal lichens (Lewis and Smith, 1967; Dahlman et al., 2003) and are well-known constituents of bacteria, fungi, and lower plants (Bielecki, 1982). The result supports the idea that fungal spores and fragments are the primary source of trehalose.”

Comment 6: Page 30783 – section 3.4: The negative vertical gradients found for the α -/ β -pinene SOA tracers are puzzling in my opinion. The authors try to come up with a reasonable explanation. However, did they consider the possibility that there could be trapping of biogenic SOA beneath the canopy? I find it quite unlikely that a large fraction of the α -/ β -pinene SOA would be produced beneath the canopy from forest floor emissions. In this respect, there is evidence for a Californian pine forest that biogenic SOA is formed just above the canopy (Holzinger et al., 2005). To support the hypothesis that a large fraction of biogenic SOA is due to forest floor emissions, measurements of α -/ β -pinene just above the forest floor would be warranted. Since such measurements are not available, the authors may want to consider trapping of biogenic SOA beneath the canopy as an

alternative explanation for their findings.

Reply 6: We agree that trapping of BSOA beneath the canopy can partly explain the negative vertical gradient. However, vertical profiles of monoterpenes reported by Holzinger et al. (2005) varied depending on local time of day: trapping of monoterpenes beneath the canopy was significant mainly at night, whereas monoterpenes were vertically well-mixed within the canopy during the day. In addition, Holzinger et al. did not report particle measurements. On the other hand, the time scale for each sample in our study is on the order of a week, which may show averaged vertical profiles of monoterpenes and others. Moreover, even when the vertical gradients of $\delta^{13}\text{C}_{\text{WSOC}}$ and potential temperature were insignificant (i.e., when vertical mixing was likely significant within the canopy), the vertical gradients of α -/ β -pinene tracers and WSOC remained significant. We believe that these profiles still support possible sources near the forest floor. Taking into account the referee's comment, we have included additional discussion of this point in the revised manuscript:

“It is also noted that trapping of WSOC beneath the canopy, which is significant mainly at night (e.g., Holzinger et al., 2005), can partly explain the negative vertical gradient. However, the time scale for each sample in our study is on the order of a week. Our data show averaged vertical profiles of WSOC including the data during the day when WSOC might be vertically well-mixed within the canopy. In addition, even when the vertical gradients of $\delta^{13}\text{C}_{\text{WSOC}}$ and potential temperature were insignificant (i.e., when vertical mixing was likely significant within the canopy), the vertical gradients of α -/ β -pinene tracers and WSOC remained significant.”

Technical corrections:

Comment 7: Page 30777 – line 1: delete “the”: show that 68% of the local

Reply 7: This has been corrected as suggested.

Comment 8: Page 30778 – line 8: I would write: . . . using a gas chromatograph (HP GC6890N, Hewlett-Packard, Palo Alto, CA, USA) equipped with a capillary column (mention here which one and the supplier) and coupled to

Reply 8: As suggested, this sentence has been rephrased as: “...using a gas chromatograph (HP GC6890N, Hewlett-Packard, Palo Alto, CA, USA) equipped with a fused silica capillary column (DB-5MS, Agilent Technologies, Santa Clara, CA, USA) and coupled to...”

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