Response to Anonymous Referee #1 comments on "*Vertical profiles of volatile organic compounds and fine particles in atmospheric air by using aerial drone with miniaturized samplers and portable devices*".

Main:

1. **Specific comment**:

It is not very clear how particle-phase VOCs were sampled, using ITEX? And how does it differ from the aerosol-phase carboxylic acid sampling using filter accessory in another method?

Author's response:

The particle-phase VOCs were collected by in-tube extraction (ITEX) system without filter accessory. An active sampler ITEX packed with sorbent material requires the pumping system for the collection of target compounds onto sorbent. This system allowed the simultaneous collection of gaseous and particle phases. Passive SPME Arrow sampling system can be employed only for the collection of gas-phase compounds due to the negligible diffusion of particle onto the coating sorbent (Peltonen and Kuljukka, 1995; Ruiz-Jimenez et al., 2019; Pusfitasari et al., 2022). The concentrations of VOCs in particle phase collected by ITEX system were calculated as substruction (differences) between the results obtained by these two sampling techniques. The sample collection and subtraction are described on lines 307-311, 475-477, and 501-502.

In the case of carboxylic acid, the samples were collected by the ITEX system furnished with filter accessory (Figures 1 – F). In this case, carboxylic acids of the aerosol particles were expected to be retained onto the filter accessory, while the gas-phase is trapped onto the ITEX's sorbent. The sample from the filter accessory was then released and the extract was analyzed by hydrophilic interaction liquid chromatography (HILIC)-MS/MS for qualitative and quantitative analyses. The sample collection is described on lines 215-219.

Changes in manuscript:

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2. Specific comment:

The authors presented gas-phase and particle-phase data separately in Figures 2-5. Is there a way to discuss more on how those VOCs partition between two phases? For instance, how do their partitioning coefficients depend on altitude and/or PM composition (BC or others).

Author's response:

In this particular study, our main focus was to develop sampling platform (an aerial drone) containing versatile miniaturized air sampling systems SPME Arrow and ITEX for reliable collection of air samples, and portable BC and CPC devices for real-time measurement of BC and PNC at high altitudes. The capability of filter accessory attached to ITEX's is also described. In addition, proper analytical measurement for VOCs/carboxylic acids by using GC-MS and HILIC-MS/MS were done by optimizing and validating the methods for the reliable results.

We understand that gas-particle partitioning of VOCs is very important to study, since it plays an important role in governing the specification, transport and deposition of the VOCs. Experimental data on equilibrium gas-particle partitioning is also essential to model descriptions of secondary organic aerosols. However, since this was not our focus in this study, we were planning to deal these items in more detail in the future manuscript.

Changes in manuscript:

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3. Specific comment:

Are the measured vertical distribution and/or temporal distribution of the VOCs/carboxylic acids depending on 1) meteorological conditions (PBL height etc.); 2) oxidative conditions (O3 levels etc.); and 3) PM concentration and characteristics (BC or PNC)?

Author's response:

In this study, the measured vertical and temporal distribution of VOCs/carboxylic acids were depending on the PBL height.

Changes in manuscript:

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Minor

1. **Specific comment**:

Figures 4-5: not sure why cumulative concentration is needed as an x-axis parameter.

Author's response:

The cumulative concentration was used to avoid misreading. For instance, in Figure 4, for hexylmethylamine in gas phase, the real concentration was 680 ng.m^{-3}, while the cumulative value was around 1500 ng.m^{-3}.

Changes in manuscript:

We have modified Figures 4-5 as separate figure by altitudes.

- Page 19, new remade Figure 4

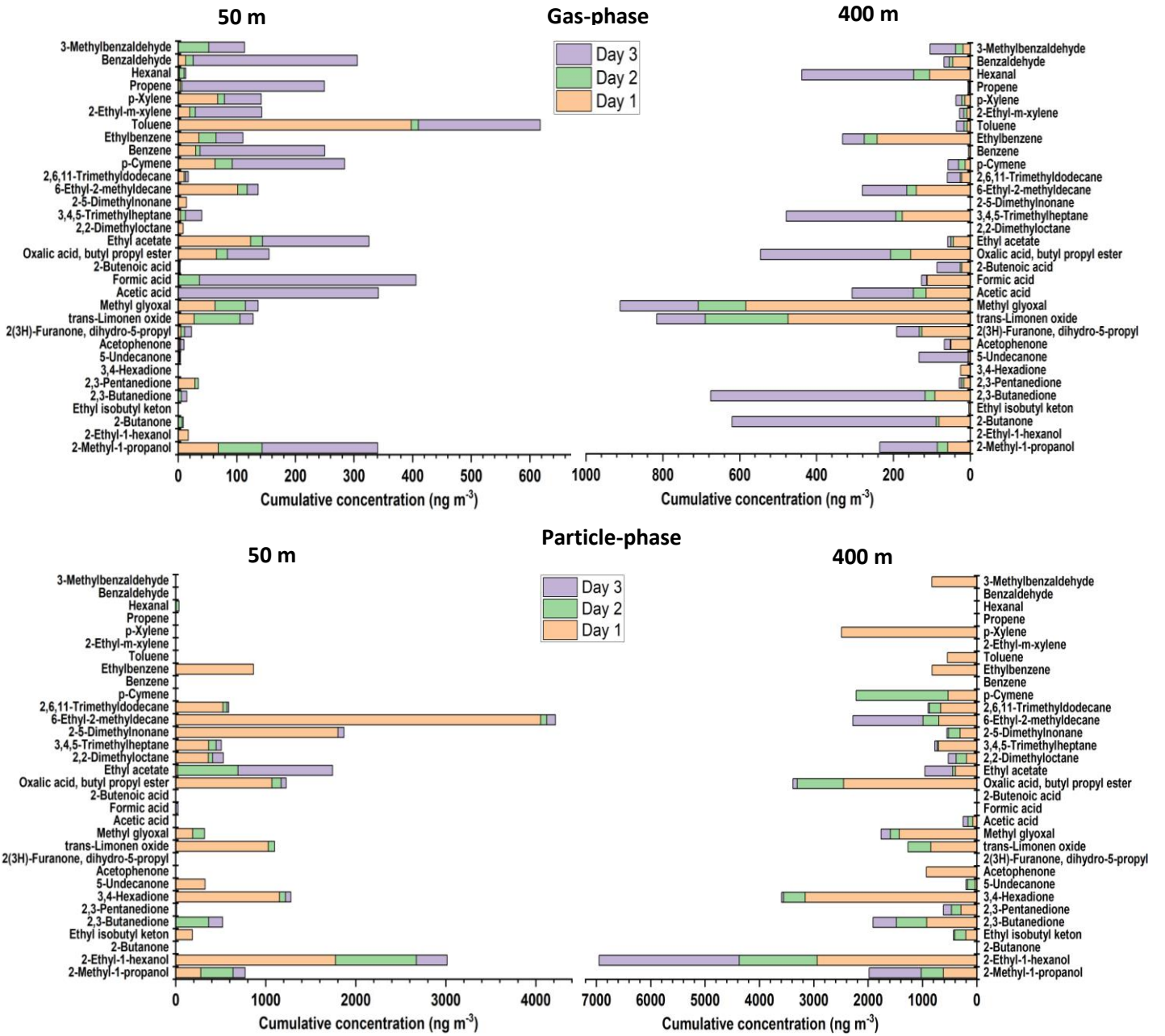
- Page 21, new remade Figure 5.

2. Specific comment

Figure 7: font size too small to see clearly.

Author's response:

We have now modified the figure with larger font size.

Changes in manuscript:

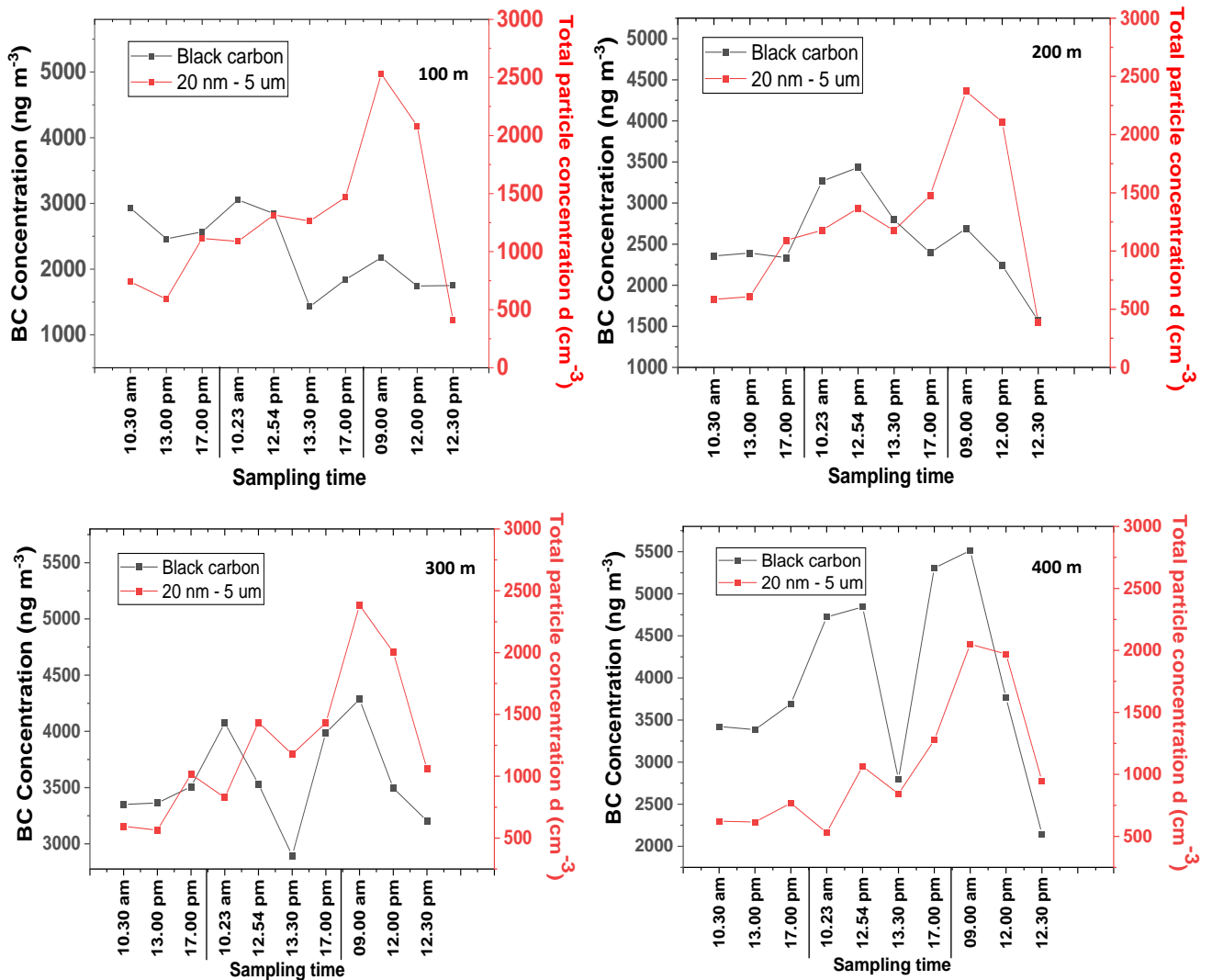
Page 26 line 598

3. Specific comment

Figure 6: how come there is such large negative concentration of BC in Phase I of Figure 6 (left)?

Author's response:

BC sensor was very sensitive for temperature and turbulence changes (Pan et al., 2011; Elomaa, 2022). When the drone started to warm up, take off, and then quickly moved vertically with the speed of 2.5 ms^{-1}, these could result in unstable and negative reading on the BC meter. When the drone approached altitude of 365 m, the BC device was stabilized just when the drone's speed slowed down.

There are also several studies agreed that the reading bias in the BC meter could be caused by the raise of amplification factor due to multiple scattering in quartz fiber matrix of the device's tape. Further, the deposition of scattering material along with BC to the filter tape produced the "shadowing effect" can cause the BC meter to show higher bias in the readings (Weingartner et al., 2003; Dumka et al., 2010).

We suspect that all these reasons caused the reading to be negative and unstable (Figure 6-left, area number I). Therefore, to further improve the reliability of the results in the future, we suggest building a portable BC monitor that includes better electronic model and to adjust better device position in the drone to reduce turbulence.

Changes in manuscript:

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Reference

Dumka, U. C., Moorthy, K. K., Kumar, R., Hegde, P., Sagar, R., Pant, P., Singh, N., and Babu, S. S.: Characteristics of aerosol black carbon mass concentration over a high altitude location in the Central Himalayas from multi-year measurements, Atmos. Res., 96, 510–521, https://doi.org/10.1016/j.atmosres.2009.12.010, 2010.

Elomaa, T.: Mustan hiilen mittaus suodatinpohjaisilla sensoreilla, University of Helsinki, 2022.

Pan, X. L., Kanaya, Y., Wang, Z. F., Liu, Y., Pochanart, P., Akimoto, H., Sun, Y. L., Dong, H. B., Li, J., Irie, H., and Takigawa, M.: Correlation of black carbon aerosol and carbon monoxide in the high-altitude environment of Mt. Huang in Eastern China, Atmos. Chem. Phys., 11, 9735–9747, https://doi.org/10.5194/acp-11-9735-2011, 2011.

Peltonen, K. and Kuljukka, T.: Air sampling and analysis of polycyclic aromatic hydrocarbons, J. Chromatogr. A, 710, 93–108, https://doi.org/10.1016/0021-9673(95)00207-4, 1995.

Pusfitasari, E. D., Ruiz-Jimenez, J., Heiskanen, I., Jussila, M., Hartonen, K., and Riekkola, M.-L.: Aerial drone furnished with miniaturized versatile air sampling systems for selective collection of nitrogen containing compounds in boreal forest, Sci. Total Environ., 808, 152011, https://doi.org/10.1016/J.SCITOTENV.2021.152011, 2022.

Ruiz-Jimenez, J., Zanca, N., Lan, H., Jussila, M., Hartonen, K., and Riekkola, M. L.: Aerial drone as a carrier for miniaturized air sampling systems, J. Chromatogr. A, 1597, 202–208, https://doi.org/10.1016/j.chroma.2019.04.009, 2019.

Weingartner, E., Saathoff, H., Schnaiter, M., Streit, N., Bitnar, B., and Baltensperger, U.: Absorption of light by soot particles: Determination of the absorption coefficient by means of aethalometers, J. Aerosol Sci., 34, 1445–1463, https://doi.org/10.1016/S0021-8502(03)00359-8, 2003.

Author response on the Anonymous Referee #2 review of "*Vertical profiles of volatile organic compounds and fine particles in atmospheric air by using aerial drone with miniaturized samplers and portable devices*" (acp-2023-40).

1. **Specific comment**:

   Line 24: Please provide the time period of the sampling.

   Author's response:

   We have now added the time period for the sampling.

   Changes in manuscript:

   Page 1, line 24.

   We modified the sentence to "In total, 135 air samples were collected under optimal sampling conditions *from October 4 to October 14, 2021* at the boreal forest SMEAR II Station, Finland."

2. **Specific comment**:

   Lines 32-33: The measurement was conducted at a rural site over the forest. Was there any BC source around the site? If not, should BC concentration be lower at the surface and higher at the high altitude? Why the results are surprising? Please clarify.

   Author's response:

   Lines 32-33 mentioned: "For spatial distribution, surprisingly the BC concentrations were increased at higher altitudes being 2278 ng m^{-3} at 100 m and 3909 ng m^{-3} at 400 m."

   There were at lower altitudes BC sources, such as living activities nearby the sampling site, including heating sauna and fuel burning from cars nearby the area. The concentrations at higher altitude were higher because of the air mixing following long-range transport from different areas.

   The results were "surprising" because the readings from the portable BC monitor were higher than those of the reference instrument (lines 553 – 554). We have addressed several factors that might contribute to these results (lines 556 – 568), such as a) the altitudes of the instruments during the measurement of the BC were different, b) electronic model of the portable BC produced the "shadowing effect" resulting in the BC meter's higher concentration values and c) the atmospheric boundary layer height (that was around 500 m in October 2021 in Hyytiälä) played an important role to govern high BC concentrations at high altitudes.

Changes in manuscript:

Page 2, line 32

In order to avoid confusion among readers, we deleted the word "surprisingly". The sentence is now becoming "For spatial distribution, the BC concentrations were increased at higher altitudes being 2278 ng m^{-3} at 100 m and 3909 ng m^{-3} at 400 m".

3. **Specific comment**:

Lines 78-87: A brief introduction of the ITEX system is required

Author's response:

Changes in manuscript:

Page 3 lines 83-86

We have now added to the text "an exhaustive sampling technique ITEX sampling system with large sorbent volume can be fully automated, and it provides continuous air sampling, reliable analysis, and quantification (Lan et al., 2019; Pusfitasari et al., 2022). As an active sampler, ITEX system allows the simultaneous collection of gas and particle phase compounds."

4. **Specific comment**:

Line 94: Was aerosol collected by ITEX tube or by filter connected to the ITEX system? Please clarify.

Author's response:

Line 94 (now it is line 96): "The compositions of different gas phase fractions, collected both by SPME Arrow and ITEX systems, aerosol particles collected by ITEX sampling including filter accessory …"

Aerosol particles were collected both by ITEX system with and without filter accessory. These were two separate experiments, while the clarifications are given in section 2.7. Shortly, lines 193-194 mentioned that ITEX sampling system without filter was exploited to simultaneously collect gas phase and particles. Whereas lines 208 – 216 described the aerosol sampling by using ITEX system furnished with filter. For the latter, the filter accessory collected aerosol particles for organic acid determination.

Changes in manuscript:

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5. **Specific comment**:

    Section 2: A table summarizing the target chemical species and their sampling and detection (including the detection limit) techniques would be helpful for the readers to understand how different species were sampled and analyzed in this study. The current presentation of the manuscript is somewhat confusing. Both the gases and particles were collected and measured by several methods.

    Author's response:

    We agree with the reviewer. We have added table below section 2.7. The limit of detections of model chemicals can be seen in Supplementary Tables S6 and S10.

    Changes in manuscript:

    Page 8 lines 198, 205, 208, and 216; we added "(Section 3.3)", "(Section 3.6)", "(Section 3.4)", and "(Section 3.5)", respectively.

    Page 9 line 222, we added "(Section 3.7)"

    Page 9 line 223, we added Table 1 below.

Table 1. Summary of target species, sampling and measurement techniques.

Target species	Sample phase	Sampler	Experiment(s)	Measurement technique
VOCs	Gas phase	ITEX + filter	Section 3.4	GC-MS
VOCs	Gas phase	SPME Arrow	Sec. 3.3; 3.4; and 3.6	GC-MS
VOCs	Particle phase	ITEX	Section 3.3 and 3.6	GC-MS
Carboxylic acids	Particle phase	Filter accessory	Section 3.5	HILIC-MS/MS
Black carbon	Particle phase	Portable AethLabs	Section 3.7	Real-time by Portable AethLabs
Total particle number	Particle phase	Portable CPC	Section 3.7	Real-time portable CPC

6. **Specific comment**:

    What was the influence of UAV propellers on VOC sampling? The sampling inlet was located underneath the UAV, under which circumstance, the sampling may be affected by UAV downwash. In addition, VOC was sampled during the descending flights. The air underneath UAV was consistently affected by the UAV propeller. Depending on the size and payload of UAV, this influence can be several 10s of meters. Then how could the sampling be representative at different heights, especially for the sampling protocols shown in Figures S1 and S4?

Author's response:

We agree with the reviewer, that UAV propellers might cause airflow disturbances during the flight and affect the concentrations of analytes in gas phase and aerosol particle in the air surrounding the UAV. The careful selection of ITEX and SPME Arrow samplers' positions on the UAV platform is needed to minimize the effect of the UAV propellers on the results during sampling.

The influence of UAV propellers and ITEX and SPME Arrow samplers' positions on VOC sampling has been investigated in our previous research (Lan et al., 2021). The effect of propellers on horizontal and vertical movements have also been studied by Jimenez et al (Ruiz-Jimenez et al., 2019). We found in this study that since the SPME Arrow system, located in the sampling box close and under the body of the UAV (i.e. our aerial drone) gave over 95% recoveries, it proved that propellers effect on the results was very minor. No significant effect was observed either on ITEX sampling system, where the recoveries ranged from 99.7 to 103.7%. Accordingly, our developed UAV with adjusted samplers' position had no significant influence on the VOC sampling.

Changes in manuscript:

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7. **Specific comment:**

Lines 151-153: If I understand correctly, the desorption processes took about 4 s (800 uL / 200 uL s-1). What were the desorption efficiency of different VOCs?

Author's response:

Desorption is critical factor to ensure maximum transfer of analytes to the GC-MS system. The optimization of parameters that influence desorption efficiency for ITEX sampler, such as injection volume, flow rate, and desorption temperature have been studied in our previous research (Pusfitasari et al., 2022). 800 uL/200 uL s^{-1} was the optimum desorption condition. The study was done using the following carefully selected VOCs as model compounds, such as diethylamine, isobutylamine, triethylamine, trimethylamine, pyridine, p-cymene, 2-butanol and 2-butanone.

Changes in manuscript:

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8. **Specific comment:**

    Lines 190-191: Please provide the detection limit of GC-MS analysis for different VOCs. Somewhere in the manuscript, the total VOC sampling volume needs to be noted. The sampling flow rate was 40 to 78 mL/min, and the sampling took about 10 min according to Figures S1 to S4. So the sampling volume ranged from 400 mL to 780 mL. The concentrations of ambient VOCs ranged from a few 100s of ng m-3 to several 1000s of ng m-3 (e.g., Figure 2). In this case, the GC-MS must be very sensitive so that it could measure VOCs with such a small amount. Please clarify.

Author's response:

The limit of the detection for selected model compounds can be found in the Supplementary Tables S6 and S10. To assure the sensitivity, we took regular GC-MS maintenance steps, such as cleaning the ion source, tuning the vacuum system, and ensuring the minimum contamination in the system since sample contamination or degradation may result in decreased MS sensitivity. Sensitivity of the GC-MS system was up to *ppt* level. Moreover, miniaturized air sampling systems ITEX and SPME Arrow that integrate sampling, isolation, and preconcentration without any solvents, used here, increased the sensitivity further.

In addition to our study, there have been other studies, where GC-MS have been utilized with similar results/sensitivity:

Lan et al (Lan et al., 2021), Pusfitasari et al (Pusfitasari et al., 2022), and Eerdekens et al (Eerdekens et al., 2009) used Agilent Technologies GC 6890A coupled to a 5973 MSD; Hakola et al employed a 5890 GC (Agilent Technology) coupled to a 5972 MSD (Hakola et al., 2012); and Hellen et al employed two GCs (Agilent Technology) coupled to 5972 MSD and 5975C MSD, respectively (Hellén et al., 2018). The MSD instruments used electron ionization (EI) as ion source.

Changes in manuscript:

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9. **Specific comment:**

Lines 227-234: Why PLSR was used for the quantification of VOC compounds? What do you mean by semi-quantification? Here list 19 standards for VOC quantification. What about the quantification of the other VOCs without standards?

Author's response:

In the environmental samples, we could detect a high number of compounds, and PLSR was developed to help the clarification, quantitation and semi quantification of the compounds. In our case, we detected up to 48 compounds. The lack of individual standards for all the identified compounds caused difficulties in the compound quantification. Then PLSR model was exploited to *semi quantify* the samples. This approach was successfully employed for semi quantification of environmental samples resulting in good prediction accuracies (with the prediction error of <16%) (Kopperi et al., 2013). For semi quantification, the data collected from the standard calibration curves were used to estimate the concentrations of the identified compounds based on PLSR of the ion intensities.

Changes in manuscript:

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10. **Specific comment:**

Line 261: Please provide the references of the "previous studies".

Author's response:

We have now added the reference. It is now line 268.

Change in manuscript:

Page 11 line 268.

"… have been tested in our previous studies (Pusfitasari et al., 2022)"

11. **Specific comment:**

Line 262: The authors mentioned that "the average sampling time that is used before reaching equilibrium for both MCM-41-Arrow and DVB/PDMS-Arrow units is about 20 minutes". However, the total sampling time was 10 min as shown in Figures S1 to S4. Please clarify.

Author's response:

The sampling time of 10 min was chosen to fit it with the maximum flight time of our drone system that was approximately 20 min including take-off and landing with single battery.

Changes in manuscript:

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12. **Specific comment**:

Figure 2: The measurement was taken over the forest. However, no information about the concentration of isoprene, monoterpenes, and sesquiterpenes was presented. Were these compounds detected? What were the concentrations of these compounds?

Author's response:

We did not see very volatile isoprene since either it might have been lost during sampling or we could not separate it by GC column of semi-polar to polar character. However, we could detect methyl glyoxal that most probably was produced in the atmosphere by the oxidation of isoprene.

From monoterpenes we observed p-cymene and oxidized limonene (i.e. trans-Limonene oxide) (see Figures 2, 3, 5, and Supplementary Table S8). Since other monoterpenes were under LOQ, they were not reported. As can be seen from Figure 5, the concentrations of methyl glyoxal and trans-limonen oxide were higher at the altitude of 400 m, while the concentration of monoterpene p-cymene was higher at lower altitude 50 m. In regards to sesquiterpenes, we could not measure them at high altitudes ≥50 m due to their highly reactive nature. Hellen et al. succeeded to measure sesquiterpenes at very low concentrations with mean concentration of 13 ppt_v at SMEAR II Station Finland, but the samples were taken from the ground level (altitude of 1.5 m). (Hellén et al., 2018)

Changes in manuscript:

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13. **Specific comment**:

Figure 3: It is difficult to believe that very volatile compounds such as benzene can present in the particle phase. Any explanation? In addition, was water vapor removed before VOC collection? How was the sampling affected by humidity, especially for the cases where a filter was presented in front of ITEX? Would water condensation increase the loss of VOCs on filter?

Author's response:

We mostly did not detect benzene in the particle phase except twice on October 9 (at 17:00, 3418 ng.m^{-3}) and October 12 (at 18:00, 455 ng.m^{-3}). We suspect that benzene in particulate phase was long-range transported from different areas. Some earlier studies have reported that carbon core of diesel exhaust particles (e.g. from buses) with large surface area contributes to the adsorption or condensation of benzene (Muzyka et al., 1998). Therefore, benzene can be found also in the particle phase (Muzyka et al., 1998; Cheng et al., 2013).

In the case of humidity concern, our PTFE filter did not have major problems due to its hydrophobic properties (Pusfitasari et al., 2022; Hou et al., 2012; Tang et al., 2019). In addition, active ITEX sampling system with proper sorbent eliminates major humidity problems. Moreover, humidity variation can be compensated by using an internal standard (Sanchez, 2014; Biagini et al., 2019). In our study, we used decafluorobiphenyl as an internal standard (lines 151-152). We did not remove water since we used short sampling time (less than 20 min), suitable ITEX sorbent furnished with filter, and internal standard.

Changes in manuscript:

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14. **Specific comment**:

Line 418: for "the first sampling system", do you mean "ITEX (+ filter accessory)"?

Author's response:

Correct.

Changes in manuscript:

Page 17 lines 426-429

Now we have modified the sentence to "In addition to the comparison of gas phase collected by ITEX furnished with filter accessory and by SPME Arrow system, the compound recoveries of gas phase obtained by the first sampling system ITEX furnished with filter were also evaluated".

15. **Specific comment:**

Lines 419-421: How was the recovery calculated?

Author's response:

We have now added the text to describe the recovery calculation.

Changes in manuscript:

Page 8 lines 212-213

The recovery was calculated from the difference between concentrations obtained by SPME Arrow and by ITEX furnished with filter accessory.

16. **Specific comment:**

Figure S6: Please use the same color code for the left and the right panels. After correction of VOC losses onto the ITEX filter, do the ITEX and SPME methods provide identical results? If not, please provide the reasons

Author's response:

Supplemental Figure S6 has now been modified to the same color. Although the results were not exactly the same, they were comparable. Slightly higher concentrations obtained by the ITEX system could be caused by higher collected amount compared to that collected by the passive SPME Arrow sampler. In addition, the sampling time was not in the equilibrium.

Changes in manuscript:

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The change was made in electronic supplementary information (ESI), Supplemental Figure S6.

17. **Specific comment:**

Section 3.5: It has been reported in the literature that there may be negative artifacts using filters to collect particles, since some of the semi-volatile organic compounds can evaporate from the filters. Please discuss this uncertainty for the filter sampling and analysis in the current study.

Author's response:

The objective of this study (section 3.5) was to show that compounds retained in filter accessory of ITEX sampling system could be analyzed. Negative artifacts that arise from the evaporation of particulate organics on the filter may cause false positive in the measurement. However, in our study we also analyzed blank filters and subtracted the concentrations achieved from the results of the natural samples, to ensure that artifacts originating from sample preparation procedure (i.e. filter extraction) were removed (Hatch et al., 2018).

Changes in manuscript:

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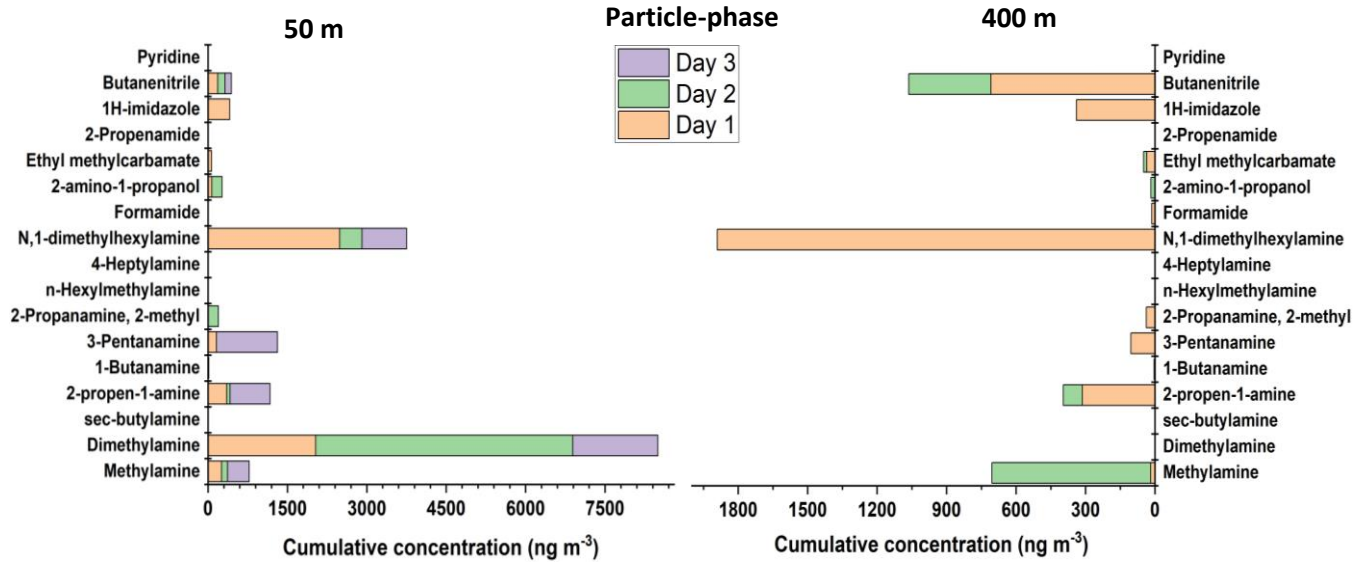
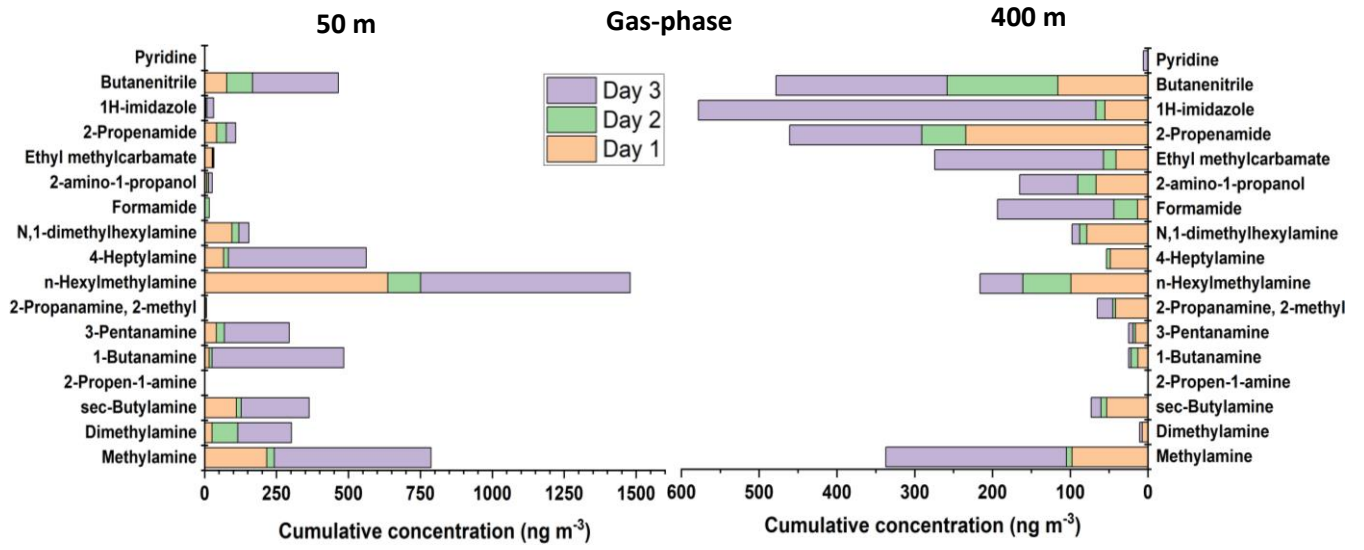
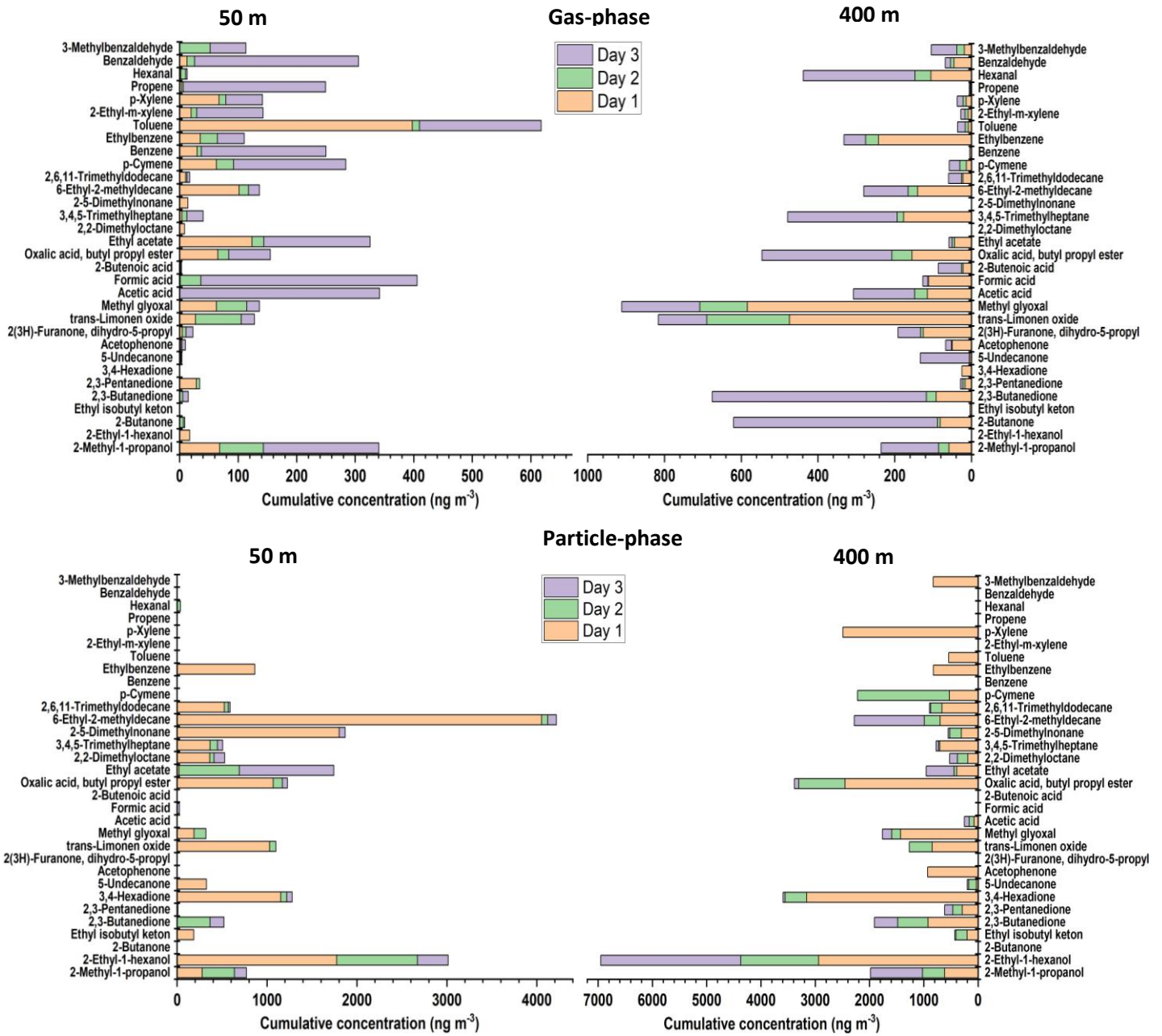
18. **Specific comment:**

Figure 5: I don't think that it is a good way to compare the concentration differences between the gas and the particle phases by adding up the results from different altitudes and different days. For example, the concentration distribution at 50 m was very different on Day 1 comparing to Day 3. Please remade the figure separating the results either by day or by altitude

Author's response:

Thank you for the critical comment related to Figure 5. Figure 4 with the same model and Figure 5 have now been modified.

Changes in manuscript:

- Page 19, new remade Figure 4.

- Page 21, new remade Figure 5.

19. **Specific comment**:

Lines 494-495: The authors mentioned that "BTX (benzene, toluene, xylene) were mostly discovered at the altitude of 50 m." BTX indicated the influence of the anthropogenic sources. This observation was different from that of BC, another anthropogenic tracer. Any explanation for this?

Author's response:

In gas phase, BTEX (benzene, toluene, ethylbenzene, and xylene) were higher at lower altitude (50 m) with the concentrations up to 410 ng m^{-3}. However, in particle-phase, BTEX were mostly detected at high altitude (400 m) with the concentrations up to 2475 ng m^{-3} (Figure 5). At lower altitudes, BTEX were emitted from gasoline (major fuel of vehicles). Our sampling took place on the yard close to the parking area that might be the reason why our sampler collected the gas phase BTEX at 50 m. This finding agreed with other study by Chen et al (Chen et al., 2018) who measured the BTX from forest canopy (at the altitude between 20 and 26 m) with the concentrations of between 100 and 300 ng m^{-3}. Since our study demonstrated that the BTEX concentrations of particle phase were higher at higher altitude (400 m), it also supported the BC observation.

Changes in manuscript:

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20. **Specific comment**:

Line 510: Change "m-3" to "m"

Author's response:

Thank you, it has now been corrected.

Changes in manuscript:

Page 22 line 521

"…the altitude between 20 and 26 m (Chen et al., 2018; Yassaa et al., 2006)"

21. **Specific comment**:

Table 2: The comparison of BC concentrations observed at the listed different locations is meaningless since the landscape, emission source, and atmospheric condition are totally different among the locations. If the authors still want to make a comparison, why not just compare the BC concentration detected over the boreal forests?

Author's response:

In this table, we tried to compare the BC concentrations at high altitude. Since we did not find BC concentrations for comparison at boreal forest at high altitude, we compared them with those of rural area found in the literature. However, we added BC concentrations of the same boreal forest, measured on the ground level (4 m).

Changes in manuscript:

Page 24 line 570

Table 3. Average BC concentrations observed at different locations.

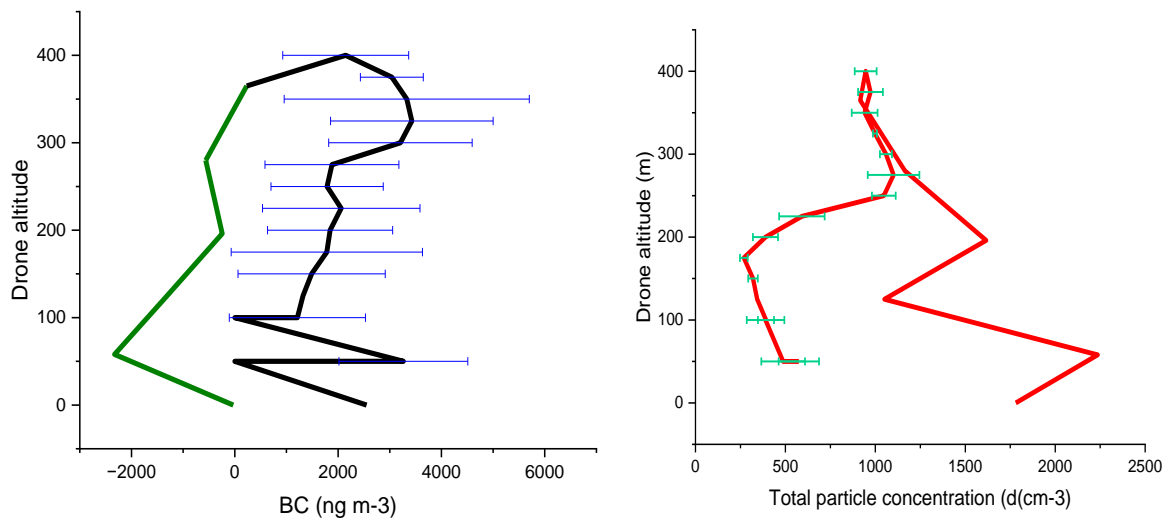
Location	Altitude	Environment	Average BC concentration (ng m^{-3})	Reference
Hyytiälä, Finland	100 m	Boreal forest	2278±1188	This study
Hyytiälä, Finland	200 m	Boreal forest	2500±1497	This study
Hyytiälä, Finland	300 m	Boreal forest	3564±1648	This study
Hyytiälä, Finland	400 m	Boreal forest	3909±729	This study
Hyytiälä, Finland	4 m	Boreal forest	320 – 1291 ± 337*	(Hyvärinen et al., 2011)
Mahabaleswar, India	1378 m	Rural	2600 ± 260	(Meena et al., 2021)
Mountain Huang, China	1840 m	Rural	1663±919	(Pan et al., 2011)
Port Blair, India	73 m	Rural	2446±66	(Moorthy and Babu, 2006)
Sinhagad, India	1300 m	Rural	1500	(Safai et al., 2007)

*320 ng m^{-3} was the annual average, while 1291 ng m^{-3} was the concentration average measured during pollution event in Autumn

22. **Specific comment:**

Figure 6: It is strange to put the altitude on the x-axis.

Author's response:

Thank you for the comment. We tried to turn the altitude to the y-axis (as below), but we found then hard to mark the horizontal movement (i.e. area III and IV as mentioned in Figure 6 of the main manuscript), and it is difficult to read the deviations due to overlapping especially when they have similar values at the same altitude. Therefore, we decided to keep the figures as they are in the original manuscript.

Changes in manuscript:

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23. **Specific comment:**

Lines 582-585: I don't think that the explanation for large BC variation is convincing. The boundary layer stratification and turbulence could affect both BC and particle number concentration measured by CPC. In addition, what was the temperature variation during the measurements when the UAV was hovering? It is hard to believe that the change in temperature would be so significant to affect BC measurement when hovering.

Author's response:

The ambient temperatures during measurement are listed in Supplementary Table S1. The temperature ranged between 7.8 – 10.2 ⁰C depending on the altitude.

Both turbulence and temperature change could affect the BC measurement results. Why this was not noticed for CPC, might be due to the different electronic device of both instruments.

BC sensor was very sensitive to temperature resulting in high variations for BC measurements, while in fact temperature did not affect much the CPC. According to Elomaa et al. (Elomaa, 2022), the continuous measurement with the temperature different at the speed of around 5 ⁰C/h, can cause the BC concentration variation of from 800 to 1000 ng/m^3. The results have also been supported by Pan et al (Pan et al., 2011) who also proved that the large variation in the BC measurements could be due to several factors such as temperature, boundary layer stratification, and turbulence. In our study, we did not adjust the position of our portable BC meter, therefore

in this manuscript we made some future suggestions regarding portable BC to be used for real-time measurement using the UAV (in the conclusion part, lines 654 – 656), such as a modified portable BC monitor should include a better electronic model and the BC device should be positioned in such a way in the UAV that it minimizes turbulence effect.

Changes in manuscript:

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Reference

Biagini, D., Lomonaco, T., Ghimenti, S., Onor, M., Bellagambi, F. G., Salvo, P., Di Francesco, F., and Fuoco, R.: Using labelled internal standards to improve needle trap micro-extraction technique prior to gas chromatography/mass spectrometry, Talanta, 200, 145–155, https://doi.org/10.1016/j.talanta.2019.03.046, 2019.

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