Comments are in black and responses in blue.

Response to Reviewer #2

The manuscript presents results from a series of well-designed experiments that explore emissions of isoprene and monoterpenes from Artic plants to the atmosphere. The experimental design is sound and well-described, except for a few minor points noted below. The main conclusion is that the rapidly warming Arctic will cause sharply increased emissions of isoprene, which previous studies have shown to have a significant impact on atmospheric chemistry. Overall, this paper supports previous research findings, but the detail and atmospheric chemistry perspective make it a valuable contribution to the literature.

Thank you for the positive feedback. Our responses to the specific comments are provided below.

While the focus of the paper is from the atmosphere-exchange perspective, several eco-physiological concepts which have been discussed previously for Arctic BVOC emissions should be addressed. First, given the emphasis placed in the paper on the response of emissions to warming, the acclimation process should be addressed (see one reference below).

That is a very good point; thank you for raising it. We have added a new subsection in the revised manuscript (Section 4.2) to discuss long-term effects of warming, including the acclimation process:

"BVOC produced by plants are involved in plant growth, reproduction, and defense, and plants use isoprene emissions as a thermotolerance mechanism (Peñuelas and Staudt, 2010; Sasaki et al., 2007). The exponential response of isoprene emissions to temperature observed at TFS adds to a growing body of evidence indicating a high isoprene-temperature response in Arctic ecosystems. However, observations at TFS do not necessarily reflect long-term effects of warming. Schollert et al., (2015) examined how long-term warming affects leaf anatomy of individual arctic plant shoots (*Betula nana*, *Cassiope tetragona*, *Empetrum hermaphroditum*, and *Salix arctica*). They found that long-term warming results in significantly thicker leaves suggesting anatomical acclimation. While the authors hypothesized that this anatomical acclimation may limit the increase of BVOC emissions at plant shoot-level, Kramshøj et al. (2016) later showed that BVOC emissions from Arctic tundra exposed to six years of experimental warming increase at both the plant shoot and ecosystem levels.

In addition to the direct impact of long-term warming on BVOC emissions, ecosystem-level emissions are expected to increase in the Arctic due to climate-driven changes in plant biomass and vegetation composition. For instance, the widespread increase in shrub abundance in the Arctic – due to a longer growing season and enhanced nutrient availability (Berner et al., 2018; Sturm et al., 2001) – will likely significantly affect the BVOC emission potential of the Arctic tundra. Additionally, as mentioned above and as discussed extensively by Peñuelas and Staudt (2010) and Loreto and Schnitlzer (2010), emissions of BVOCs might be largely beneficial for plants, conferring them higher protection from abiotic stressors which are predicted to be more severe in the future. Long-term arctic warming may thus favor BVOC-emitting species even further."

Second, temperature in the current study refers to air temperature, but isoprene and other MT emissions respond to leaf temperature. And leaf temperature in turn depends on plant water relations in addition to air temperature. Given the unique eco-hydrology of tundra plants, some attention should be paid to this driver. In particular, was soil moisture monitored for any of the chamber experiments? Was SM measured at the tower?

We unfortunately did not monitor soil moisture but have made it clearer that the current study refers to air temperature:
"Over the course of the two field campaigns at TFS, BVOC surface emission rates were measured over a large span of enclosure temperatures (2-41°C). While isoprene and MT emissions respond to leaf temperature (Guenther et al., 1993), air temperature was used here in place of leaf temperature – which has been assumed before in the literature for high-latitude ecosystems (e.g., Olofsson et al., 2005; Potosnak et al., 2013). Several studies have, however, suggested a decoupling of leaf and air temperature in tundra environments (Lindwall et al., 2016; Potosnak et al., 2013). With predicted increase of air temperature in the Arctic, it still remains largely unknown how leaf temperature will change and impact BVOC emissions. As suggested by Tang et al. (2016), long-term parallel observations of both leaf and air temperature are needed. The response of BVOC emissions to temperature discussed here should be interpreted with this potential caveat in mind."

Line 62: Delete second "to."

Done, thanks for noticing this typo.

Line 81: The term "flanks" is a bit odd. At least it should be singular.

Done.

Line 86: Italicize "Vaccinium vitis-idaea."

Done.

Line 109: very briefly give the details on the "moisture trap." Were cooled glass beads used?

We have added a short description of the moisture trap in the revised manuscript: "The moisture trap was a U-shaped SilcoSteel™ tube (stainless steel treated) cooled using thermoelectric coolers". Note that the tube was empty (no glass beads).

Line 110: What absorbents were used?

This has been added to the revised manuscript: "Analytes were concentrated on a Peltier-cooled multistage micro-adsorbent trap (50 % Tenax-GR and 50 % Carboxen 1016)".

Line 129: How large was this combined effect, in percent terms?

We observed a progressive 80 % decline in CFC-113 peak area.

Line 154: What uncertainty is introduced by data processing? Do you mean something related to statistics?

The error introduced by data processing relates to the error in averaging the data from 2 minutes to 10 minutes, as well as error induced from peak fitting in the data processing software.

Line 166: Please note when solar noon occurs at the site in AST.

This has been added to the revised manuscript: "A total of eight vertical profiles were performed at ~3-hour intervals between 12:30 pm AST on June 15, 2019 and 11:00 am AST on June 16, 2019 in order to capture a full diurnal cycle (solar noon around 2 pm AST)".

Line 179: Add a brief mention of how the tubes were capped.

This has been added to the revised manuscript: "Following collection, adsorbent cartridges were sealed with Teflon-coated brass caps and stored in the dark at ~4°C until chemical analysis".

Lines 315-318: I concur with this conclusion. You could make this more clear and impactful by stating that both the intense wildfires regionally and the isoprene emissions locally were driven by high air temperature. But further, could there have been an influence on the photochemical lifetime of isoprene due to the products of the wildfire? Could the main isoprene oxidants, OH & ozone, be suppressed?

The occurrence of wildfires depends on meteorology (e.g., temperature and soil moisture) but also vegetation type and coverage, and lightning frequency. Fire emissions are a complicated mixture of trace gases and aerosols, many of which are short-lived and chemically reactive. This mixture affects the atmospheric composition in complex ways that are not completely understood. Recent measurements during the NASA/NOAA ATOM and FIREX-AQ campaigns have shown that wildfires might actually be responsible for increased ozone mixing ratios in aged plumes (Bourgeois et al., in prep). Our surface ozone measurements at Toolik Field Station suggest that mean ozone mixing ratios increased from ~27 ppb during June 1-19, 2019 to ~34 ppb during the June 20, 2019 fire event.

Lines 319-335: Since you are integrating results and discussion, what's the implication of these results?

We have added the following sentence in the revised version of the manuscript: "This record of ambient air isoprene, MACR, and MVK mixing ratios is, to the best of our knowledge, the first in an Arctic tundra environment. The combined measurement of isoprene and its oxidation products provides a new set of observations to further constrain isoprene chemistry under low-NO_x conditions in atmospheric models (Bates and Jacob, 2019)."

Lines 338: There is no need to refer explicitly back to the Materials and Methods section, so "(see Section 2.3)" can be removed.

Done.

Lines 343-345: What's the implication? Is the isoprene 'sticking around' from the more productive part of the day or is production continuing throughout the 'night' (low-PAR conditions).

We now refer to Section 3.2.2: "Samples collected on June 16, 2019 from 4 to 4:30 am (see Fig. 5f) show decreasing isoprene mixing ratios with increasing elevation, suggesting higher levels (25-50 pptv) in the nocturnal boundary layer than above. This result suggests continuing isoprene emissions by the surrounding vegetation under low-PAR conditions. This is further discussed in Section 3.2.2".

Lines 347-350: This is more than 'consistent.' I would change the wording to something along the lines of 'expected.'

This has been modified in the revised manuscript: "This maximum at ground-level is expected for a VOC with a surface source (Helmig et al., 1998) while the 200 pptv mixing ratio can likely be attributed to a temperature-driven increase of isoprene emissions by the surrounding vegetation."

Line 383: this is a _really_ high number and should be highlighted in the abstract. Unadjusted for temperature, biomass and light, it's similar to results from many midland low-latitude forests. Later in the paragraph, you give the comparison, which is good. But, I think the key is that the extreme values are so high.

We agree and have highlighted this result in the abstract of the revised manuscript.

Line 406-409: Should explicitly state that even with nearly 24 hours of light, still get the typical diurnal pattern. The key is that low sun angles translate to very low PAR (non-linearly), and therefore you still see the typical diurnal pattern. Later in the paragraph, you get to this explicitly, but the discussion should be combined for clarity. Also, this should be related to the diurnal balloon experiment results.

We have clarified this in the revised manuscript: "Figures 8a-c show the mean diurnal cycle (over the two campaigns) of isoprene surface emission rates for different vegetation types. The two field campaigns were carried out during the midnight sun period, which could possibly sustain BVOC emissions during nighttime. It should, however, be noted that low sun angles translate to very low PAR and a typical diurnal pattern is observed in summer at TFS despite 24 hours of light (see Fig.8h)."

We have also related these results to the balloon vertical profiles: "These sustained BVOC emissions during nighttime confirm observations by Lindwall et al. (2015) during a 24-hour experiment with five different Arctic vegetation communities and explain the higher isoprene levels observed in the nocturnal boundary layer than above during the diurnal balloon experiment (see Section 3.1.2)."

Lines 455-459: This needs to be tempered a bit. There are issues of timescales and acclimation. Also, I assume there are relatively few chamber measurements between 35 and 40 deg C, hence a leveling off is within statistical probabilities. Also, you will argue against this point in the following paragraph, so this could be presented more clearly to readers.

We have added the number of chamber measurements in each temperature bin in the revised Figure 9. We do agree that, given the relatively few chamber measurements at T > 30°C, a leveling-off is within statistical probabilities. We have therefore tempered this paragraph accordingly in the revised manuscript (and in the abstract):

"While the model predicts a leveling-off of emissions at approximately 30-35°C, our observations reveal no such phenomenon within the 0-40°C enclosure temperature range (Fig. 9). However, given the limited number of enclosure measurements above 30°C, a leveling-off of emissions cannot be statistically ruled out. The key result here is that MEGAN2.1 adequately reproduces the temperature dependence response of Arctic ecosystems in the 0-30°C temperature range – ambient temperature > 30°C being unlikely."

Lines 461-462: I think I understand what this sentence is trying to convey, but it is confusing and the statement could be clearer. You that for every year in the dataset, there were 1-23 days with a temp above 20 deg C?

We have clarified this sentence in the revised manuscript: "Additionally, for each year in the 1988-2019 historical dataset, there were only 1 to 23 days (0 to 4 days) per year with a maximum temperature above 20°C (above 25°C)".

Line 471: "Under" might be a better word chance than "scarcely."

The wording has been changed accordingly in the revised manuscript.

Line 472: Same comment as Line 81 about "flanks."

Done.

Line 474: "Elevated" compared to what? Expectations or previous measurements?

This has been clarified in the revised manuscript: "While the overall mean isoprene emission rate amounted to 85 μgC/m^2/h at TFS, elevated (> 500 μgC/m^2/h) isoprene surface emission rates were observed for *Salix* spp., a known isoprene emitter."

Line 477: Thermotolerance hasn't been addressed previously in the manuscript. At the minimum, a citation is necessary, but it might be best to remove this if it's not explored more thoroughly with regards to Arctic plants.

Thermotolerance is now addressed in Section 4.2 of the revised manuscript (see above).

Line 485: Can remove "likely" since "suggesting" is already in the sentence and provides sufficient caution.

Done.

Line 486-490: Here thermotolerance is addressed a bit further, with references. But it would be better to have a short paragraph or group of sentences that speculates specifically about the role thermotolerance could play in promoting isoprene-emitting species in the Artic. The current allusions here and at Line 477 makes the topic appear as tacked on.

This is now addressed in Section 4.2 of the revised manuscript (see above).

Line 839, Table 1: some mention of the lack of measurements for R. chamaemorus would be useful, since it is the dominant species.

Indeed. This has been added in the revised manuscript: "The tundra vegetation around TFS is heterogeneous but most dominant species (except *Rubus chamaemorus*) were sampled."

Line 992, Figure 5: Solid, colored lines connecting the points would help visually highlight vertical trends.

We have updated this Figure in the revised manuscript (see below; one panel per balloon flight) to make it easier to distinguish measurement points at different heights.

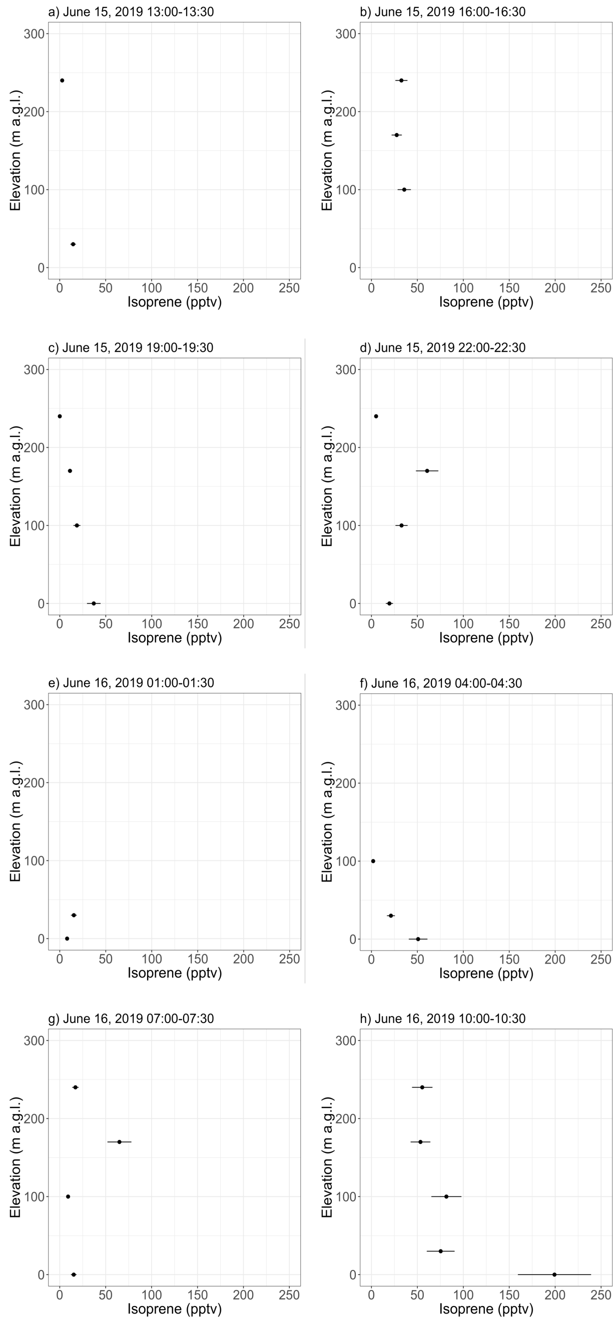

Figure 5: Vertical profiles of isoprene mixing ratios as inferred from 30-min samples collected with a tethered balloon. The error bars show the analytical uncertainty for isoprene (20 %). Samples with an isoprene mixing ratio lower than blanks were discarded. Hours are in Alaska Standard Time (UTC-9).