

Here are the authors' responses to the reviewers' comments. The comments are shown in bold font, the responses are shown in red and the related modifications of manuscript are shown in blue.

Response to Reviewer 1:

General Comment

The authors use a 1-D model to study the trend of OH, NO₃ and H₂SO₄ in boreal forest over 2007-2018 at SMEAR II, Finland. They compared their model results to observations at SMEAR II station. They show that their model agrees with some observations during 2007-2018, but inconsistent with others. They then use their model to calculate the long-term trend of OH, NO₃ and H₂SO₄. Finally, they also compared the modeled values of OH, NO₃ and H₂SO₄ to some proxies. The paper is trying to address a very interesting question, about the long-term trend of oxidants in boreal forest in southern Finland.

We really appreciate the reviewer for the valuable comments. Our replies to the comments are given below.

Major Comment

1. I am a bit of concerned about the long-term trend in this study. As the authors showed in Figure 4, their BVOC does not really agree with observations. If I look at Figure 4, I see a decrease of observed monoterpene concentrations from 2007 to 2018. In contrast, their model calculations in Table 1 show an increase of 50% of monoterpene emissions for the same period. As monoterpene is presumably the most important species to determine OH, NO₃ and H₂SO₄ in this work, I am not sure how robust these long-term trends are. Can the authors plot the observational trend of monoterpene concentrations from Figure 4? If the observational trend is opposite to the model calculation on monoterpenes, there seems to be little value to discuss the model trends of oxidants in this paper.

The reviewer is right about the measured monoterpene concentration trend, which is decreasing from 2007 to 2018 (here "to 2018" means until the end of 2018, the same below), and the modelled concentration is increasing. However, considering the measurement gaps, especially the gap between 2014 to 2016, we need to compare them with more accurate methods and get a closer look of the trends. The measurement data we used here were from five measurement heights: 32 m, 54 m, 74 m, 101 m, and 125 m. The model data were from all the levels between 32 m and 120 m.

Here we only consider the model data points when measurements were available to calculate the linear fitting. First, if we separate the datasets according to the measurement gap between 2014 to 2016, we can get two periods before and after the gap, one is from 2007 to 2013 and the other is from 2017 to 2018. The linear fittings show that the modelled trends of these two periods are $-0.72 \times 10^8 \text{ molec cm}^{-3} \text{ yr}^{-1}$ and $19 \times 10^8 \text{ molec cm}^{-3} \text{ yr}^{-1}$, and the measured trends are $-6.8 \times 10^8 \text{ molec cm}^{-3} \text{ yr}^{-1}$ and $6.5 \times 10^8 \text{ molec cm}^{-3} \text{ yr}^{-1}$, respectively (Fig. R1). Therefore, both the model and the measurement show a decreasing trend from 2007 to 2013 and an increasing trend from 2017 to 2018. The lower trends of measurement data may result from several very low measurement concentrations around the beginning of 2012 and 2018 (Fig. R1).

Secondly, the one-year moving averages of modelled and measured monoterpene concentrations also show consistent variations, both of which have shown a consecutive strong peak and dip from 2010 to 2014, and a sharp increase from 2017 to 2018 (Fig. R2).

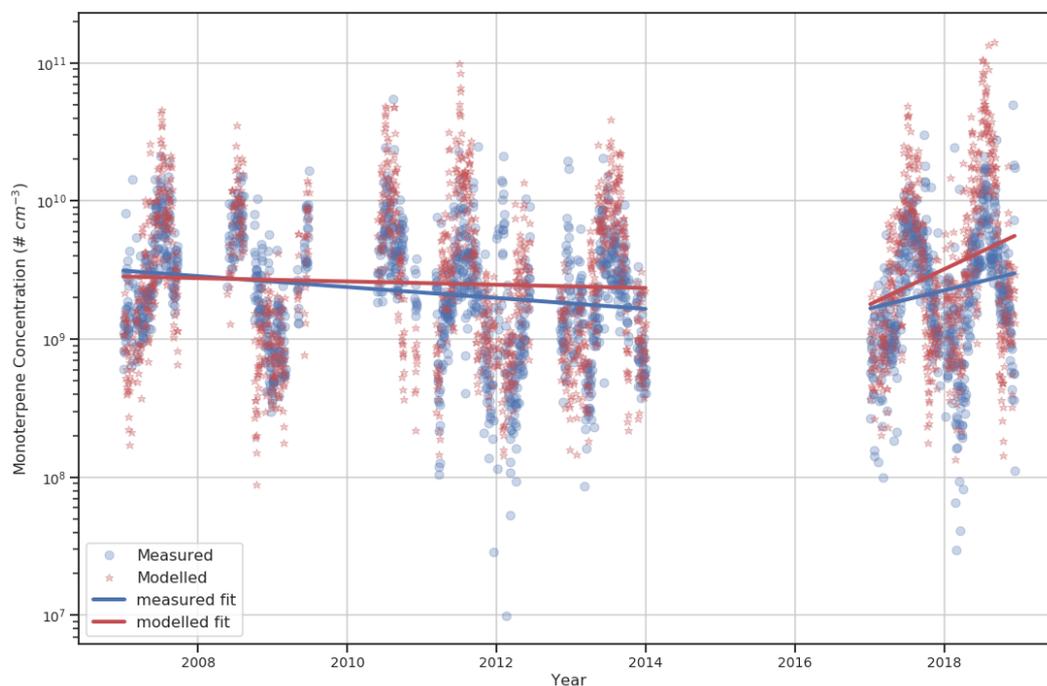


Figure R1: Time series of modelled (red star) and measured (blue circle) monoterpene concentrations from 2007 to 2018. The linear fits of the logarithmic data are plotted as solid lines in the same color for modelled and measured data. The modelled data points are plotted only when measured data are available.

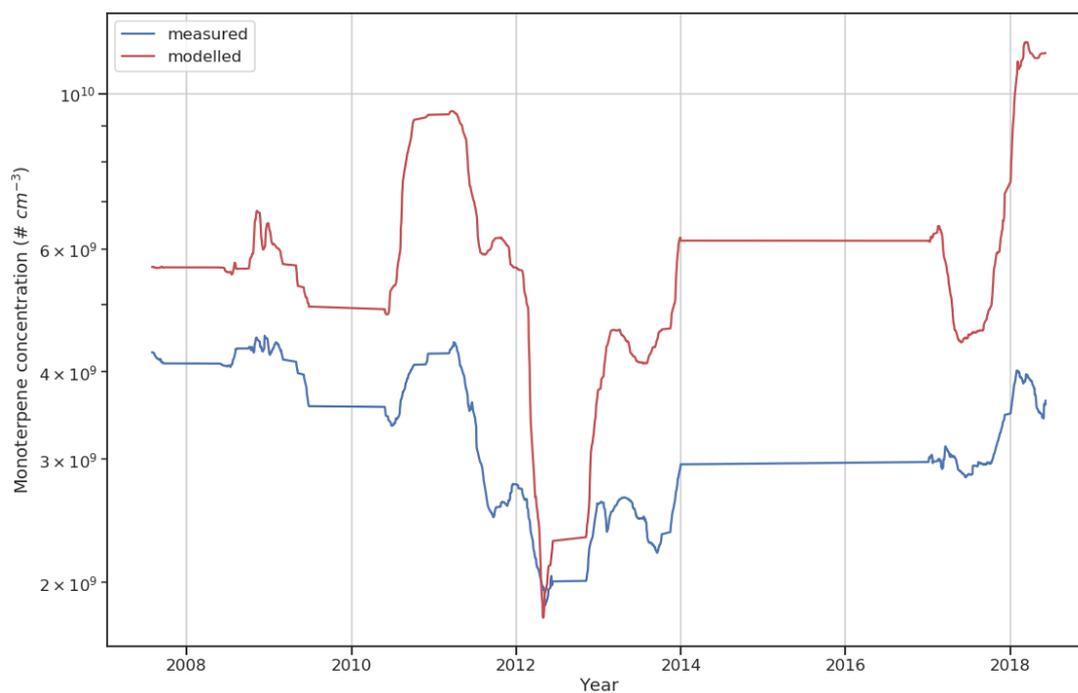


Figure R2: The 1-year running mean of modelled (red) and measured (blue) monoterpene concentrations from 2007 to 2018.

We will add Figs. R1 and R2 to the supplemental material as Figs. S6 and S7, and the text in the manuscript will be modified as shown below:

P15, L389: "... measured data points."

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"... measured data points.

In order to compare the modelled and measured trends of monoterpene concentrations, we only consider the model data points when measurements were available to calculate the linear fittings or the trends. First, considering the measurement gap during 2014 to 2016, we can separate the datasets into two periods before and after the gap. One is from 2007 to 2013 and the other is from 2017 to 2018. The linear fittings show that the modelled trends of these two periods are -0.72×10^8 molec $\text{cm}^{-3} \text{yr}^{-1}$ and 19×10^8 molec $\text{cm}^{-3} \text{yr}^{-1}$, and the measured trends are -6.8×10^8 molec $\text{cm}^{-3} \text{yr}^{-1}$ and 6.5×10^8 molec $\text{cm}^{-3} \text{yr}^{-1}$, respectively (Fig. S6). Therefore, the trends in the model and the measurement agree well, both of which show a decreasing trend from 2007 to 2013 and an increasing trend from 2017 to 2018. Secondly, the one-year moving averages of modelled and measured monoterpene concentrations also show consistent variations, both of which have shown a consecutive strong peak and dip from 2010 to 2014, and a sharp increase from 2017 to 2018 (Fig. S7). The lower trends of measurement data may result from several very low measurement concentrations around the beginning of 2012 and 2018 (Fig. S7)."

P16, L392:

"... 2018 for the height interval 20 m-40 m."

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"... 2018 for the height interval 32 m - 125 m. The measurement median data were from the mean values of five measurement heights: 32 m, 54 m, 74 m, 101 m, and 125 m. The model median data were calculated from the mean of all the levels between 32 m and 120 m."

I understand that there is some agreement on OH and H₂SO₄ between model and measurements (Figure 5 and Figure 8), but those snapshot agreements do not really help to constrain the long-term trend of those oxidants. Some observational constraints on the long-term trend of biogenic emissions would be critical for this paper.

We agree that the OH observation data are not long enough to represent the whole period due to the challenging field measurements of OH. Therefore, this is also a motive of this study in which we aim to provide some understanding of long-term OH concentrations at this site, especially when measurements were not available. As for H₂SO₄, we believe that the long-term observations with the CI-API-TOF from 2016 to 2018 are one of the longest H₂SO₄ measurement available globally. Therefore, the comparison using this dataset is our best option currently. Furthermore, the model results show a good agreement with measurements from 2016 to 2018 (Figs. 8 and S5). Thus we assume that the model is able to be used to support the long-term analysis of H₂SO₄ in this study.

Unfortunately there were no long-term measurements of emission rates of BVOCs, however, the measured and estimated emission data in previous studies have already been analyzed and used to constrain the modelled BVOC emissions in SOSAA (Zhou et al. 2017a). Moreover, in this study the modelled and measured monoterpene (the largest contributor of OH reactivity at SMEAR II among BVOCs, Mogensen et al., 2015) concentrations (Figs. 4, R1 and R2) and fluxes (Fig. S3) agree well. Accordingly, the modelled monoterpene emissions provided by the MEGAN module in SOSAA can be considered to be able to constrain OH in our simulations.

2. Model values of oxidants used in this paper. After reading the paper a few times, I am still unclear which model level was used to provide the data for the long-term trend in Table 1 and elsewhere. Is this the average between 0 and 150 m, 20 and 40 m, or other levels? How do the authors make sure that the levels they chose are representative of the whole vertical domain? I would strongly suggest that the authors make it clear that what model level was used for the data and why.

We thank the reviewer to remind us about the confusion. Indeed, we used different levels for different data. We try to make them clearer here. We will add one extra column to show the height levels in Table 1 (see below).

Actually the aim of our study is not to represent the whole vertical domain, but focus inside the surface layer. We didn't use the data inside the canopy, because of two reasons. First, the mixing chemical compounds are not homogeneous as above the canopy, and we want to avoid this uncertainty. Secondly, the measurements that we used to compare with are generally placed in open ground without vegetation, which is more similar to the atmospheric condition above the canopy.

The new table is shown below and the modified part are labelled as red. Note that we recalculated the values for monoterpenes with new height interval (between 32 and 120 m) to make it consistent with the analysis in the manuscript. The previous height interval used in Table 1 is between 0 and 120 m.

Parameter	Height levels	Median (Mean*)	Yearly trend	P _{MK}	12 years trend
SWR*	125 m	259.5 [W m ⁻²]	0.616 (-2.767; 3.332) [W m ⁻²]	0.162	7.39 [W m ⁻²]
TEMP*	mean of 4.2, 8.4, 16.8, 33.6, 50.4 and 67.2 m; added 125 m after August 2012	278.0 [K]	0.07 (-0.005; 0.214) [K]	0.769	0.80 [K]
		279.4 [K]	0.07 (-0.004; 0.215) [K]	0.740	0.80 [K]
		277.3 [K]	0.07 (-0.011; 0.206) [K]	0.815	0.79 [K]
RH*	mean of 4.2, 8.4, 16.8, 33.6, 50.4 and 74.0 m; added 101 and 125 m after February 2014	80.2 [%]	-0.53 (-1.412; 0.156) [%]	0.215	-6.36 [%]
		77.1 [%]	-0.54 (-1.359; 0.179) [%]	0.206	-6.49 [%]
		81.4 [%]	-0.51 (-1.280; 0.156) [%]	0.233	-6.11 [%]
O ₃	mean of 4.2, 8.4, 16.8, 33.6, 50.4 and 74.0 m; added 101 and 125 m after February 2014	7.9E+11 [# cm ⁻³]	-0.050 (-0.62; 0.25) %	0.566	-0.60 %
		8.0E+11 [# cm ⁻³]	-0.173 (-0.71; 0.11) %	0.877	-2.01 %
		7.6E+11 [# cm ⁻³]	-0.027 (-0.65; 0.40) %	0.426	-0.32 %
CO	mean of 4.2, 8.4, 16.8, 33.6 and 50.4 m; added 101 and 125 m after February 2014	3.8E+12 [# cm ⁻³]	-0.483 (-1.80; 0.36) %	0.516	-5.65 %
		3.8E+12 [# cm ⁻³]	-0.515 (-1.78; 0.37) %	0.498	-6.01 %
		3.9E+12 [# cm ⁻³]	-0.471 (-1.99; 0.37) %	0.503	-5.51 %
SO ₂ *	16.8 m	4.8E+9 [# cm ⁻³]	-2.384 (-3.94; -1.55) %	0.012	-25.14 %
		5.4E+9 [# cm ⁻³]	-1.929 (-3.43; -0.85) %	0.022	-20.84 %
		5.0E+9 [# cm ⁻³]	-2.398 (-3.89; -1.48) %	0.013	-25.27 %
CS	ground level	4E-3 [s ⁻¹]	-1.414 (-5.33; 1.32) %	0.524	-15.71 %
		4E-3 [s ⁻¹]	-1.735 (-5.53; 0.50) %	0.440	-18.94 %
		5E-3 [s ⁻¹]	-1.251 (-5.10; 1.64) %	0.632	-14.02 %
EM-MON	mean of model layers between 2 and 20 m	1.7E+6 [# cm ⁻³]	0.773 (0.07; 2.13) %	0.158	9.68 %
		2.5E+6 [# cm ⁻³]	0.665 (0.01; 1.72) %	0.214	8.28 %
		1.4E+6 [# cm ⁻³]	0.641 (-0.05; 1.98) %	0.197	7.97 %
MON	mean of model layers between 32 and 120 m	5.2E+9 [# cm ⁻³]	2.951 (0.76; 4.66) %	0.008	36.38 %
		5.2E+9 [# cm ⁻³]	3.001 (0.67; 5.53) %	0.019	42.59 %
		5.7E+9 [# cm ⁻³]	2.619 (0.76; 5.26) %	0.007	41.76 %
OH	mean of model layers between 20 and 40 m	3.8E+5 [# cm ⁻³]	2.435 (0.61; 1.70) %	0.034	33.47 %
		1.3E+6 [# cm ⁻³]	1.560 (0.02; 3.47) %	0.104	20.41 %
		2.3E+5 [# cm ⁻³]	2.786 (1.72; 4.54) %	0.032	39.06 %
HO ₂	mean of model layers between	5.1E+7 [# cm ⁻³]	3.572 (2.41; 4.89) %	0.013	52.37 %

	20 and 40 m	1.2E+8 [# cm⁻³]	2.878 (2.09; 4.10) %	0.041	40.56 %
		3.2E+7 [# cm⁻³]	3.664 (2.38; 5.19) %	0.012	54.01 %
H₂SO₄	mean of model layers between 20 and 40 m	2.5E+5 [# cm⁻³]	-3.363 (-8.66; 0.72) %	0.377	-33.67 %
		1.2E+6 [# cm⁻³]	-5.115 (-11.12; 0.39) %	0.207	-46.74 %
		1.6E+5 [# cm⁻³]	-3.128 (-8.28; 0.80) %	0.342	-31.71 %
NO*	mean of 4.2, 8.4, 16.8, 33.6 and 50.4 m; added 101 and 125 m after February 2014	1.1E+9 [# cm ⁻³]	-0.037 (-0.33; 0.14) %	0.857	-0.44 %
		3.1E+9 [# cm ⁻³]	-0.228 (-0.78; 0.09) %	0.885	-2.70 %
		1.0E+9 [# cm ⁻³]	0.022 (-0.08; 0.10) %	0.655	0.26 %
NO₂	mean of 4.2, 8.4, 16.8, 33.6 and 50.4 m; added 101 and 125 m after February 2014	2.5E+10 [# cm ⁻³]	-1.580 (-2.60; -0.96) %	0.032	-17.40 %
		2.6E+10 [# cm ⁻³]	-3.163 (-5.54; -1.70) %	0.076	-32.00 %
		2.9E+10 [# cm ⁻³]	-3.908 (-6.06; -2.66) %	0.025	-38.02 %
N₂O₅	mean of model layers between 20 and 40 m	1.1E+8 [# cm⁻³]	-8.253 (-11.91; -6.18) %	0.013	-64.43 %
		6.6E+7 [# cm⁻³]	-6.656 (-10.12; -4.75) %	0.017	-56.24 %
		1.8E+8 [# cm⁻³]	-8.821 (-12.30; -6.81) %	0.010	-66.98 %
NO₃	mean of model layers between 20 and 40 m	3.8E+6 [# cm⁻³]	-3.518 (-4.86; -2.38) %	0.011	-34.93 %
		2.8E+6 [# cm⁻³]	-2.463 (-4.03; -0.98) %	0.028	-25.86 %
		6.2E+6 [# cm⁻³]	-3.921 (-5.10; -2.88) %	0.009	-38.12 %

Table 1 caption will be modified as:

P9, L267-273:

"Table 1: Median or mean values, yearly and 12-year trend (in percent or absolute value), PMK values and in brackets the confidence interval of the trend for the yearly change (SWR = short wave irradiance, TEMP = temperature, RH = relative humidity, CS = condensational sink, EM-MON = emission rate for monoterpenes, MON = concentrations of monoterpenes). The first, second and third rows for each variable except SWR represent daily, daytime and nighttime values, respectively. Modelled results are presented in bold. Detailed description for periods selected (daily, daytime and nighttime) are provided in the text. Statistical methods are explained in subsection 2.3."

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"Table 1: Median or mean values, height levels, yearly and 12-year trend (in percent or absolute values), and PMK values of different parameters. The units of absolute values are put inside square brackets. The confidence intervals of the yearly trends are written inside the brackets in the "Yearly trend" column. The first and second numbers show the 5th and 95th percentiles of the yearly trend slopes obtained from 1000 bootstrapping iterations, respectively. The first, second and third rows for each parameter except SWR represent daily, daytime and nighttime values, respectively. Modelled results are presented in bold. Detailed statistical methods are explained in section 2.3. The chemical compounds are written in their chemical formulae except MON which represents monoterpenes. The acronyms of other parameters are: SWR = short wave irradiance, TEMP = temperature, RH = relative humidity, CS = condensation sink, EM-MON = emission rate for monoterpenes."

The height related text in manuscript will be modified as shown below:

P18, L458-459: For EUCAARI, the exact measurement height is unknown, Petäjä et al. (2009) mentioned that it was close to ground level. The model height level is at 32.8 m to represent OH concentration above canopy. For HUMPPA-COPEC, the measurement height was at ground level outside the white container in a forest clearing at SMEAR II (Williams et al., 2011), the model height level is at 32.8 m.

P19, L469: "Modelled OH concentrations for the years 2007 to 2018 ..." --> "Modelled OH concentrations between 20 m and 40 m for the years 2007 to 2018 ..."

P20, L500: "Modelled NO₃ concentrations for the years 2007 to 2018 ..." --> "Modelled NO₃ concentrations between 20 m and 40 m for the years 2007 to 2018 ..."

P22, L527: "Measured and modelled daily median sulphuric acid concentrations ..." --> "Measured (35 m) and modelled (32.8 m) daily median sulphuric acid concentrations ..."

P23, L542: "Modelled sulphuric acid concentrations for the years 2007 to 2018 ..." --> "Modelled sulphuric acid concentrations between 20 m and 40 m for the years 2007 to 2018 ..."

P25, L590: "Yearly mean daily time series of OH concentrations estimated by SOSAA (Model) ..." --> "Yearly mean daily time series of OH concentrations estimated by SOSAA (Model) at 32.8 m ..."

P26, L608: "Yearly mean daily time series of H₂SO₄ concentrations estimated by SOSAA (Model) ..." --> "Yearly mean daily time series of H₂SO₄ concentrations estimated by SOSAA (Model) at 32.8 m ..."

P27, L626: "Yearly mean daily time series of NO₃ concentrations estimated by SOSAA (Model) ..." --> "Yearly mean daily time series of NO₃ concentrations estimated by SOSAA (Model) at 32.8 m ..."

3. OH trend explained by CO. I do not quite follow the explanations about OH trend. If OH increase by 2.8%/yr, how can this be explained by the trend of CO, which decreases at 0.5%/yr? Note that CO only accounts for 40% of OH reactivity.

In order to make the relationship between OH and CO more clear, we analyzed the trends of OH and CO in different seasons separately. Figure R3 shows the seasonal median values of OH and CO from 2007 to 2018 for four seasons. In spring and summer, OH concentrations varied slightly and did not show apparent trends, although CO showed large interannual variations especially from 2014 to 2018. This indicates that in spring and summer when OH and BVOCs concentrations are high, CO accounts for a less amount of OH reactivity, especially in summer when CO had the lowest concentrations compared to other seasons.

In autumn, OH had two peaks with the values of $0.98 \times 10^6 \text{ # cm}^{-3}$ in 2013 and $1.23 \times 10^6 \text{ # cm}^{-3}$ in 2015, which corresponded to the two dips of CO concentrations (Fig. R3). However, in autumns of 2010, 2011 and 2016, OH was not correlated with CO closely as in other years. In 2011 and 2016 the reason could be that the low CO concentrations accounted for much less OH reactivity. In 2010, other factors may play an important role which need further study. In winter when CO concentration was highest among all the seasons, the OH trend was mostly negatively correlated with CO except in 2011. Therefore, in winter, CO was the main sink of OH and CO concentration is able to explain the OH trend.

From Fig. 6 in the manuscript, we can see that the long term increasing trend of OH mainly results from the higher concentrations from 2013 to 2017, which is consistent with the OH trends in autumn and winter (Fig. R3). Therefore, as a dominant sink of OH in winter, CO is the main factor to explain the long term OH trend.

We will add the following explanation to the paper and put Fig. R3 to supplemental material as Fig. S8.

P20, Line 491: In order to investigate the relationship between OH and CO more clearly, we analysed the trends of these two compounds in different seasons separately. Figure S8 shows the seasonal median values of OH and CO from 2007 to 2018 for four seasons. In spring and summer, OH concentrations varied slightly and did not show apparent trends, although CO showed large inter-annual variations especially from 2014 to 2018. This indicates that in spring and summer when OH and BVOCs concentrations are high, CO only has a marginal impact on the OH concentrations compared with the first and higher order reactions of OH with organic compounds. On the contrary, in winter when CO concentrations were highest among all the seasons, OH trend was mostly negatively correlated with CO except in 2011. Therefore, in winter, CO is the main sink of OH and CO concentration is able to explain the OH trend. From Figures 6 and S8, we conclude that the long term increasing trend of OH mainly results from the higher concentrations from 2013 to 2017, which is consistent with the OH trends in autumn and winter. Therefore, as a dominant sink of OH in winter, CO is the main factor to explain the long term OH trend.

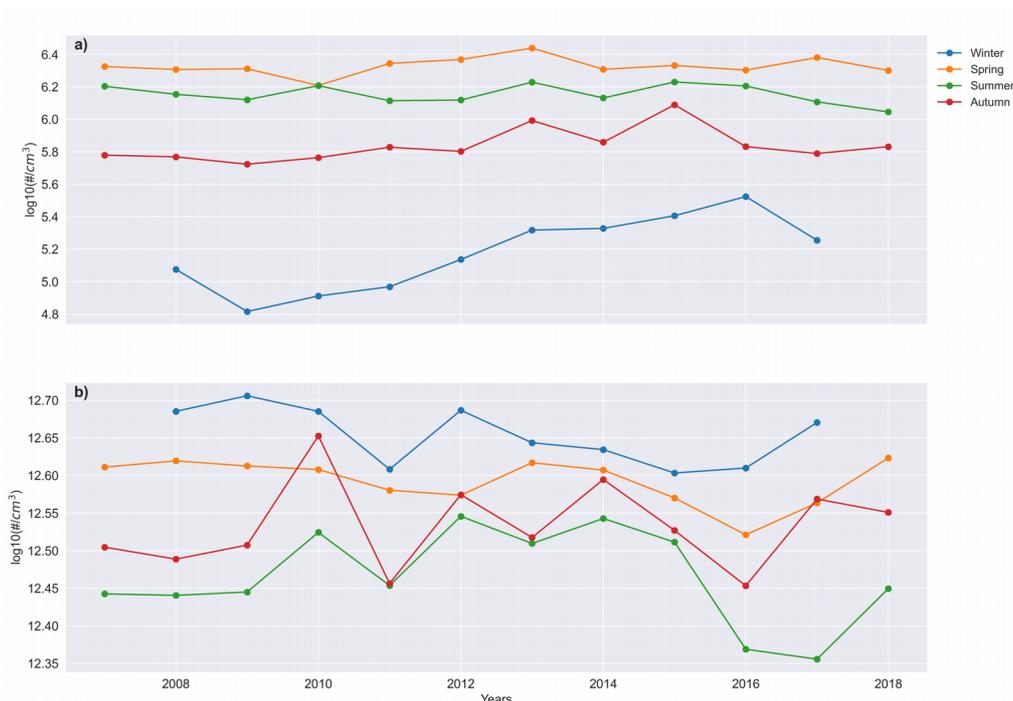


Figure R3: Seasonal medians of daytime (a) OH and (b) CO concentrations in logarithm scale for winter (blue), spring (orange), summer (green) and autumn (red) from 2007 to 2018.

4. What is the possible reason for the declining trend of NO₃ and H₂SO₄ in their model?

The main source of NO₃ comes from the oxidation of NO₂ by O₃. In Table 1 we can see that nighttime O₃ only shows a very slight decrease of less than 1% during 12 years, while nighttime NO₂ shows a 38% decreasing. The results are consistent with the decreasing trend of nighttime NO₃ which is also about 38% from 2007 to 2018. Therefore, the declining trend of NO₃ can be totally explained by the decrease of NO₂ during nighttime.

The trend of H₂SO₄, as generally analyzed in section 3.5, is greatly influenced by the large inter-annual variations. The decrease trend could mainly result from the dramatic drop in 2016 (Fig. 9), which can be explained by the decreasing of SO₂ during 2016 and 2017 (Fig. S2). We should notice that at the same time in 2016, OH concentration is higher than median, and the trend of OH shows

an increase of about 33% from 2007 to 2018. It indicates that H₂SO₄ is mainly modulated by SO₂, and OH is not a limitation in the production process of H₂SO₄.

Furthermore, the decreasing trends of the trace gas precursors NO_x and SO₂ mainly result from the environmental policies of European Community and others during last decades. For example, IMO (International Maritime Organization) regulations changed so that after the year 2015 the maximum allowed sulphur content in the shipping fuel (% in mass) used in the Baltic and the North Sea was decreased from 1.0 % to 0.1%

([http://www.imo.org/en/OurWork/Environment/PollutionPrevention/AirPollution/Pages/Sulphur-oxides-\(SOx\)-%E2%80%93-Regulation-14.aspx](http://www.imo.org/en/OurWork/Environment/PollutionPrevention/AirPollution/Pages/Sulphur-oxides-(SOx)-%E2%80%93-Regulation-14.aspx)).

The manuscript will be modified accordingly as shown below

P21, L508: "... the decreasing trend at daytime (-2.46% yr⁻¹)."

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"... the decreasing trend at daytime (-2.46% yr⁻¹). The 12-year trend of nighttime NO₃ can be explained by the decreasing of nighttime NO₂, since the main source of NO₃ comes from the oxidation of NO₂ by O₃. In table 1 we can see that nighttime O₃ only shows a very slight decrease of less than 1% during 12 years. However, the nighttime NO₂ shows a reduction of 38%, being consistent with the decreasing trend of nighttime NO₃ which is also about 38% from 2007 to 2018."

5. I am not sure how the proxy comparison in Section 3.6 is related to the trend in this work.

Probably the title has caused some misleading of the aim of this study. Actually one aim of this study is to fill the measurement gaps and to provide the long-term datasets of OH, NO₃ and H₂SO₄ at SMEAR II. These datasets can thus act as a background of different important trace gases at this site, which could facilitate the following research. Before this study, the proxies were quite commonly used to do a similar job for these trace gases, i.e., filling the measurement gaps, providing a hint of the trends, and sometimes being used as databases. The comparison between our complex process-based model and the simplified proxies aims to provide a reference of the evaluation for both of them.

Therefore the title will be modified as:

"The trend of the oxidants in boreal forest over 2007-2018: comprehensive modelling study with long-term measurements at SMEAR II, Finland"

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"Comprehensive modelling study of OH, NO₃ and H₂SO₄ over 2007 - 2018 at SMEAR II Finland with long-term measurements"

Technical edits:

1. Table 1, please explain what is the brackets for Yearly trend? What is the first number and the second number for?

In Table 1 the values in the brackets in the 'Yearly trend' column show the confidence interval of the yearly trend. They are calculated by the bootstrapping method described in Section 2.3. So here the first number and the second numbers show the 5th and 95th percentiles of the yearly trend slopes obtained from 1000 bootstrapping iterations, respectively.

2. Line 234-238, what is the criteria for Mann-Kendall test? Can the author provide some context here?

We'd like to thank reviewer for showing interest in the statistical approach of this study. Here we have tried to identify possible trends in our data and explain them without going out of the scope.

In the manuscript, we have showed the results of our Mann-Kendall test. Mann-Kendall test is a rank-based nonparametric test often applied to a series of observations in order to find out underlying trends. It is a nonparametric test meaning it works for all distributions regardless of if the dataset meets the assumption of normality or not. This test has been widely used to evaluate trends in various kinds of atmospheric data (e.g. meteorological variables and chemical concentrations) (Mitchell et al., 1996; Yunling et al., 2005; Sicard et al., 2007; Quinn et al., 2009). For our 12 years long-term data we wanted to give a conservative estimate of the monotonicity of the long-term trend. This gives a clue of how steady the long-term change of a variable. For example, a couple of exceptionally warm summers in the 12-year period have a big influence on monoterpene emissions and its trend, and extrapolating the future concentrations of monoterpenes based on that trend would be risky. Monotonicity was primarily tested with the bootstrapping method, which was explained in 2.3. In addition to this method, we have provided a second estimate of the monotonicity of the trend using the Mann-Kendall correlated seasonal test. The Mann-Kendall test assumes that a monotonic trend, if it exists, is in the same direction in all seasons.

We would also like to mention that we have applied different statistical methods to our dataset, however, we have opted to go with Mann-Kendall because it is widely used in atmospheric trend studies and it is relatively easier to understand without needing to have deep statistical knowledge.

3. Line 241, incomplete sentence.

The sentence will be modified as:

P8, L241: "The average trendumber field. You see the Edit Fields dialog. shown in time series plots is ..." --> "The trend lines shown in time series plots are ..."

Response to Reviewer 2:

General Comment

Chen et al. presented an inter-comparison study between a 1D model framework and an observational dataset from the SMEAR II site for a decade. The research site is well documented by a number of previous publications. The attempt to compiling 10 year worth of the dataset is obviously quite ambitious thus potentially beneficial to the community. Although the motivation is sound, the scientific merit of presented discussion on the observed data and the model outcomes is not well followed up. The main reason for this in my mind is that manuscript simply contains too much of information to be considered as one paper. In this sense, if the authors' main research interests are in elucidating oxidants at the site,I would recommend to just focus on the topic. Therefore, I will present my comments on the oxidant section (3.4).

We really appreciate the reviewer for the valuable comments. We agree that this paper should put more highlight on the main focus. Our aim is to show the temporal variations of OH, NO₃ and H₂SO₄ over period of 12 years. The reason to insist on including H₂SO₄ is because it plays a crucial role in aerosol processes at this site. We will reply to the comments below and decided to change the title of the manuscript accordingly (see below).

3.4.1. Ozone The discussion in the current form is not acceptable to be published. Ozone is not like other oxidants such as OH and NO₃ presented in this manuscript as it is difficult to simulate using a 1D model due to its long lifetime. In this sense, I would recommend quantitatively analyze the impacts of vertical distribution of monoterpenes to ozone distributions as Kurpius and Goldstein (2003, GRL 10.1029/2002GL016785) presented.

We thank the reviewer for the suggestion. Kurpius and Goldstein (2003) provided a comprehensive analysis of the sink terms of O₃ in a ponderosa pine forest. It showed that chemistry is the dominant daytime O₃ sink within the canopy, especially during summer when chemistry can contribute about 51%. Zhou et al. (2017b) also analyzed O₃ sink terms in a similar way with the same model SOSAA within the canopy at this site but only in one month (August, 2010), and they found that chemical removal of O₃ only contributed little to the total O₃ uptake within the canopy. Therefore, in this study we did not focus on the analysis of the chemical production and removal of O₃. Moreover, instead of calculating O₃ concentrations as in Zhou et al. (2017b), in our simulations the measured vertically averaged mixing ratios of O₃ were used as the input values for all the model layers (Section 2.2). This could also help to overcome the difficulty of simulating O₃ with a 1D model for a long-term simulation.

Here considering the negligible trend of O₃ (Table 1) and the reviewer's general comments that this manuscript contains too much information, we decide to remove the Section 3.4.1. And we will also modify the manuscript title to only focus on the analysis of OH, NO₃ and H₂SO₄.

The manuscript title will be modified as:

"The trend of the oxidants in boreal forest over 2007-2018: comprehensive modelling study with long-term measurements at SMEAR II, Finland"

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"Comprehensive modelling study of OH, NO₃ and H₂SO₄ over 2007 - 2018 at SMEAR II Finland with long-term measurements"

OH Again the presented discussion is not quantitative enough. It comes to a surprise the attempt to explain long term trend of OH by correlating with the CO trend. Considering the abundance of reactive BVOCs, I would not expect that CO would determine the presence of OH. This section is required to include a detailed description on chemical mechanisms to elucidate the long term trend of OH and its determining factor.

We'd like to thank the reviewer for bringing attention to this topic. Reviewer 1 also commented on the explanation of long term OH trend in his/her third question in the major comment, we made further explanation and replied there. Please see the response to reviewer 1 question 3 in the major comment.

NO₃ I would recommend more conducting more work on model evaluations. I understand there has not been any observational attempt of NO₃ in the research site. However, for the

model evaluation perspective, the authors can certainly use other previous community field campaign datasets open to public. Without the evaluation, it is very difficult to evaluate the validity of the discussion in this section.

The NO₃ radical has a very short lifetime which makes the direct measurement of NO₃ challenging, especially in a low NO_x environment. At SMEAR II, Liebmann et al. (2018) tried to measure NO₃ directly but its mixing ratios were always below the detection limit (1.3 pptv) during the whole campaign (05.09.2016 - 21.09.2016). At the same time, Liebmann et al. (2018) measured the NO₃ reactivity based on which they estimated the NO₃ mixing ratios. In Fig. R4 the modelled NO₃ mixing ratio during the same time period as in Liebmann et al. (2018) is plotted to be compared to the results in Fig. 10 of Liebmann et al. (2018) (shown here as Fig. R5). Our results show very low NO₃ mixing ratios which are also always below 1.3 pptv. Compared to their calculated stationary state NO₃ mixing ratios, our results are generally lower but in the same order of magnitude. Moreover, both results show the peaks on 7th, 10th, 15th, 21st of September. Therefore, our simulations of NO₃ can be considered as reasonable.

We will add Fig. R4 to the supplemental material as Fig. S9.

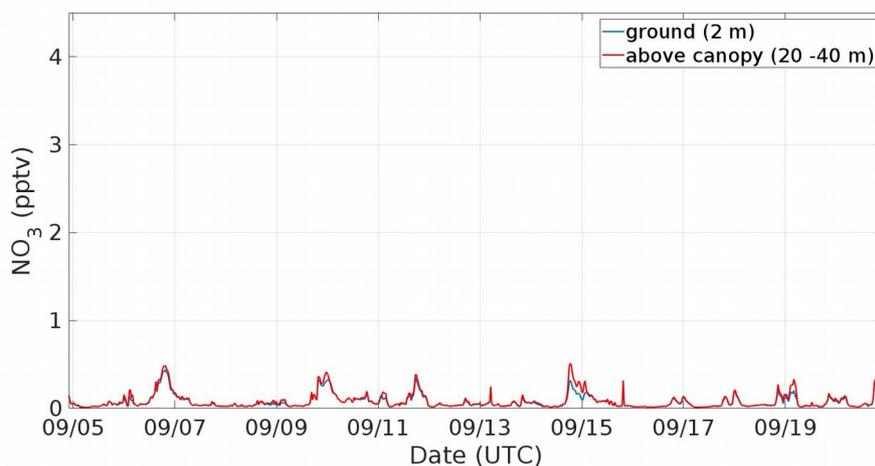


Figure R4: NO₃ mixing ratio in the unit of pptv at the ground level (blue) and above the canopy (red) from 05.09.2016 to 21.09.2016. The y limit is set to be the same as that in Fig. 10 in Liebmann et al. (2018).

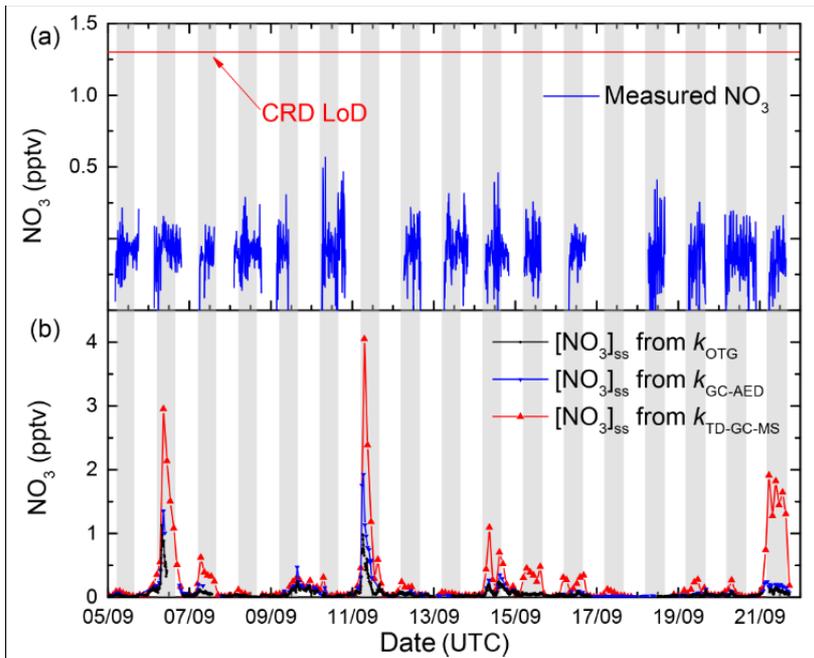


Figure 10. Stationary state NO₃ mixing ratios calculated from the production term ($k_1[\text{NO}_2][\text{O}_3]$) and using either $k_{\text{OTG}} + k_2[\text{NO}] + J_{\text{NO}_3}$ (**(b)**, black line), $k_{\text{GC-MS}} + k_2[\text{NO}] + J_{\text{NO}_3}$ (**(b)**, red line), or $k_{\text{GC-MS}} + k_2[\text{NO}] + J_{\text{NO}_3}$ (**(b)**, blue line) as loss terms. For comparison, the measured NO₃ mixing ratios are also plotted (**(a)**, blue line) as well as the 1.3 pptv limit of detection (horizontal red line).

Figure R5: Copied from Figure 10 in Liebmann et al. (2018).

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Others

1. We checked the manuscript and will change all SF --> S to make it consistent.

P21, L506: "SF2" --> "Fig. S2"

P21, L523: "SF5" --> "Fig. S5"

P23, L550: "SF2" --> "Fig. S2"

2. Typos:

P17, L438: "COPECC-HUMPPA in 2010" --> "HUMPPA-COPEC in 2010"

P18, L459: "EURCAARI (A) and the COPPEC-HUMPPA (B) campaigns." --> "EUCAARI (A) and the HUMPPA-COPEC (B) campaigns."