

We thank the referee for the time and effort to review the manuscript and appreciate the insightful thoughts and comments. Below we respond to all the concerns and suggestions, and the manuscript will be revised accordingly.

We have highlighted our responses in blue with the referee's original comments in black.

RC1: 'Comment on acp-2022-20', Anonymous Referee #1, 04 May 2022

1. In the study concentration levels of PAHs, sugars and OPAHs in PM₁₀ were studied at one urban background and three residential areas in Sweden and results were compared to the modelling results. Population exposure was also estimated, which was interesting and showed the impact of the study more clearly. Sampling and analytical methods were well-described and manuscript is well-written. There have been some earlier publications on PAHs and wood combustion from Nordic countries, but there is very little knowledge on the OPAHs. Therefore results on OPAHs could be discussed more. In addition to this, I had only minor comments:

Response: We agree with the referee that the OPAH results could be further discussed to better understand atmospheric OPAHs. We greatly appreciate the referee's comment and will revise "Section 3.3" accordingly as below.

3.3 Impact of non-residential biomass burning and OPAHs

The measured levels of PAHs, OPAHs and sugars in airborne PM₁₀ taken at the urban background (TK) around the Walpurgis Night (Apr 30) in 2016 are shown in Fig. 6. A clear elevation of both the total sugar and levoglucosan can be observed in the sample collected on the event day. In the present study, the total sugar level showed an increase of almost three times, from 36.4 to 117 ng m⁻³. The total OPAH level was also shown to increase approximately threefold. The sugar ratios (4.4-7.2) from the urban background indicated a mixed emission source of hard and soft wood combustion, except for the sample collected on the 30th Apr. This sample exhibited sugar ratio of 8.8, i.e. reflecting hard wood burning. Detailed information is given in Table S5.

The dominating OPAH was 7H-benzo[de]anthracen-7-one (benzanthrone, BAQ), accounting for more than 50% of the measured OPAHs (55 - 76%), followed by 9,10-anthracenedione (AQ) (4 - 28%) (Fig. xx). The same trend has been reported in atmospheric PM_{2.5}, measured at an urban background site in Bologna, Italy (Pietrogrande et al., 2022). The correlation between OPAH and its parent PAH was significant ($p < 0.01$), indicating PAHs and OPAHs were from the same primary combustion source or PAHs were the secondary source of OPAHs. However, the increased emission of OPAHs on the Walpurgis Night is mainly from the primary combustion source, i.e. biomass burning (Fig. xx).

Pietrogrande, M.C., Bacco, D., Demaria, G., Russo., M, Scotto., F. and Trentini, A: Polycyclic aromatic hydrocarbons and their oxygenated derivatives in urban aerosol: levels, chemical profiles, and contribution to PM_{2.5} oxidative potential, Environ. Sci. Pollut. Res., doi.org/10.1007/s11356-021-16858-z, 2022.

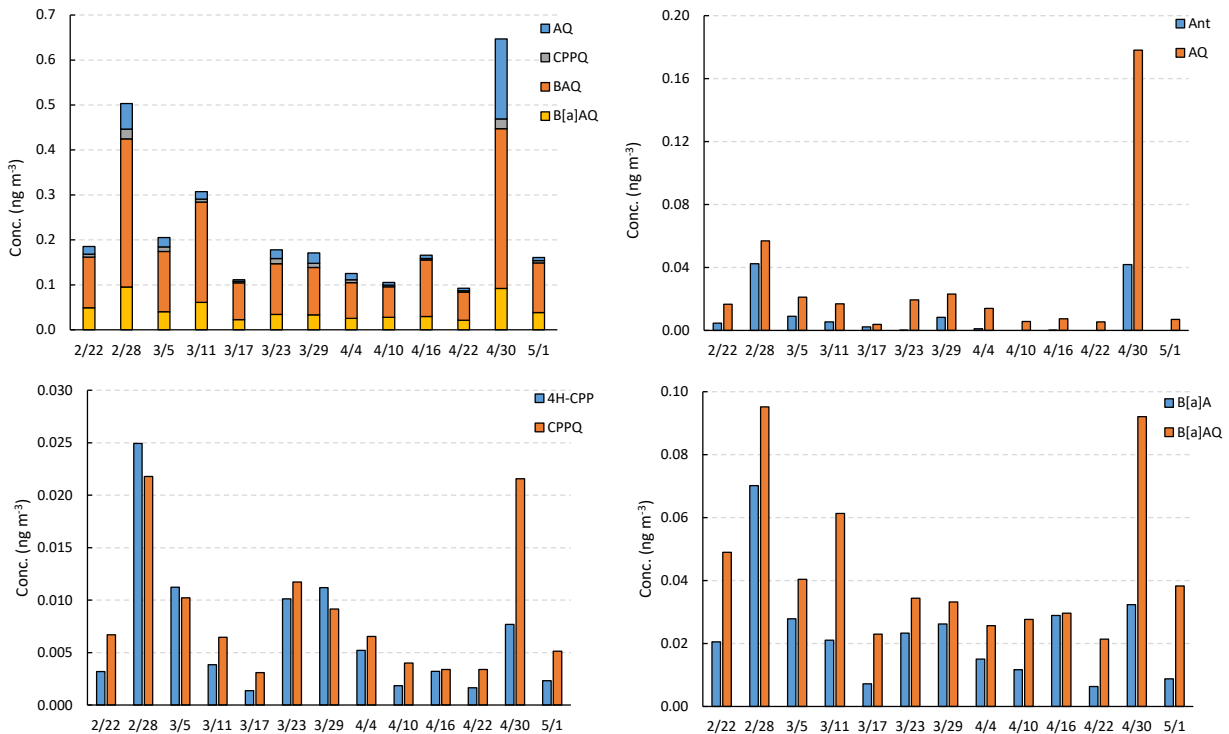


Figure xx. Measured concentration of total OPAHs (AQ, CPPQ, BAQ, and B[a]AQ) and individual concentration of OPAH and its parent PAH in PM₁₀ collected from Stockholm urban background (TK) during Feb 22 – May 5, 2016.

2. Page 2, line 30: Lifetime of levoglucosan has been estimated to be 1.8 days on average. So it is not that long living (Li, Y., Fu, T-M., Yu, J.Z., Feng, X., Zhang, L., Chen, J., Suresh Kumar Reddy, B., Kawamura, K., Fu, P., Yang, X., Zhu, L., and Zeng, Z.: Impacts of Chemical Degradation on the Global Budget of Atmospheric Levoglucosan and Its Use As a Biomass Burning Tracer, Environmental Science & Technology, 55, 5525-5536, <https://doi.org/10.1021/acs.est.0c07313>, 2021).

Response: We thank the referee that there are other studies indicating the atmospheric degradation of levoglucosan during a short period than previously reported. We greatly appreciate the referee's comment and will revise the text accordingly as below.

"A highly selective tracer for burning of wood is levoglucosan (1,6-anhydro-β-D-glucopyranose), a monosaccharide derivative formed when cellulose is pyrolysed at high temperatures (Shafizadeh, 1968; Simoneit et al., 1999). Due to the high concentration in the smoke and a high chemical stability, this tracer compound can be detected in the atmosphere through long-range transports (Simoneit et al., 1999; Fraser and Lakshmanan, 2000). In addition, mannosan and galactosan released from the thermal degradation of hemicellulose are also detected in wood smoke emissions and suggested to be source-specific tracers for wood burning (Nolte et al., 2001; Simoneit, 2002). However, recent studies indicate that atmospheric degradation of levoglucosan is important and may shorten the lifetime significantly (Li et al., 2021). They suggest considering the aging in air masses of levoglucosan when it is used to calculate the biomass burning contribution to organic

[carbon \(Hong et al., 2022\). In our case the sites were located in close proximity to the main sources of levoglucosan and the photochemical degradation is expected to be insignificant for these conditions."](#)

Li, Y., Fu, T.-M., Yu, J.Z., Feng, X., Zhang, L., Chen, J., B, S.K.R., Kawamura, K., Fu, P., Yang, X., Zhu, L. and Zeng, Z.: Impacts of chemical degradation on the global budget of atmospheric levoglucosan and its use as a biomass burning tracer, *Environ. Sci. Technol.*, 55 (8), 5525-5536, doi: 10.1021/acs.est.0c07313, 2021.

Hong, Y., Cao, F., Fan, M.-Y., Lin, Y.-C., Gul, C., Yu, M., Wu, X., Zhai, X. and Zhang, Y.-L.: Impacts of chemical degradation of levoglucosan on quantifying biomass burning contribution to carbonaceous aerosols: A case study in Northeast China, *Sci. Total Environ.*, 819, 152007, doi.org/10.1016/j.scitotenv.2021.152007, 2022.

3. Page 3, lines 1-3: Where? In this study or earlier studies?

Response: We understand that it was not clearly written in the manuscript where they were measured. It referred to previous studies measured in various cities of central and northern European countries. We greatly appreciate the referee's comment and will revise the text accordingly as below.

"Earlier studies have measured either levoglucosan, or the combination of all three monosaccharides, to estimate the contribution of wood burning to the total air PM collected in various cities of central and northern European countries (Yttri et al., 2005; Caseiro et al., 2009; Maenhaut et al., 2012, 2016; Wagener et al., 2012; Glasius et al., 2018)."

4. Maybe you could mention somewhere that some of the PAHs you studied are semivolatile and significant fraction of them may be found in gas phase.

Response: We agree with the referee, and this will be added in the section "3.1 Seasonal variation of PAH and sugar levels. PAHs". The revised text will be as below.

"The PAH concentrations showed a strong seasonal variation at all three sites as shown in Fig. 3. On the left side, the mass concentration of low and high molecular weight PAHs (LMW and HMW) are compared together with those of the total PAHs. 11 PAHs with three and four rings were grouped in LMW while 22 five- and six-ring PAHs were in HMW. [PAHs are semi-volatile compounds and partitioned in gas and particle phases depending on the volatility. Three- and four-ring PAHs are distributed mostly in the gas phase during warmer season and vice versa \(Bi et al., 2003\). The same applied to this study where LMW PAH levels were lower during summer due to their partitioning more in the gas phase.](#) The pie chart (right side in Fig. 3) shows the relative abundance of LMW and HMW during summer (Jun-Jul) and winter (Jan-Feb). A considerable shift in the relative LMW level from winter to summer was observed in all three locations with the largest change in DE. In addition, the distinctive seasonal shift observed in DE was from the increased residential heating, which mostly affected the PAHs with four rings. The increased emission of four-ring PAHs from domestic

heating was also reported in the high Arctic during winter (Singh et al., 2017). The sugar levels, in general, followed the seasonal variation as the PAHs, however, there were occasions with increased sugar levels when biomass burning or wood combustion happened.”

Bi, X., Sheng, G., Peng, P., Chen, Y., Zhang, Z., and Fu, J.: Distribution of particulate- and vapor-phase n-alkanes and polycyclic aromatic hydrocarbons in urban atmosphere of Guangzhou, China, *Atmos. Environ.*, 37 (2), 289-298, doi:10.1016/S1352-2310(02)00832-4, 2003.

5. Could you also comment somewhere how close to the EU annual limit values/thresholds you were?

Response: We agree with the referee that it could be mentioned how the measurement in this study was in relation to the EU annual limit. The comparison between the measured and EU target annual mean B[σ]P concentrations are made and will be inserted in the section “3.2 PAH and sugar emission trend associated with residential wood burning” as below.

“The annual mean concentration of B[σ]P from the three sites (DE, EN, YJ) and one rural background (ASP) were 0.11, 0.08, 0.09, and 0.06 ng m⁻³, respectively (Table S4), which is far below than the EU target value of 1 ng m⁻³.”

6. Page 8, lines 22-24: Could it also be that during summer time more of these 3 and 4 ring PAHs partition to the gas phase?

Response: We agree with the referee, and this will be added in the section “3.1 Seasonal variation of PAH and sugar levels. PAHs”. Please see the revised text above (Response to comment # 4).

7. Figure 4: Which are the locations if fig a)-c)?

Response: We appreciate the referee’s comment that the information of the location is missing in Fig. 4. The figure legend will be revised as below.

“Figure 4. Distribution of annual measurement data for levoglucosan, mannosan, and galactosan from three sampling locations. Mannosan and galactosan are determined as sum due to co-elution in the chromatogram: **(a)** DE, **(b)** EN and **(c)** YJ.”

8. Figure 8: For EN and YJ local wood and traffic has same pattern in the figure.

Response: We appreciate the referee’s comment and will change the data format to differentiate four data series as below.

