Response to RC2: 'Comment on acp-2022-20', Anonymous Referee #2, 05 May 2022

See author response in blue as below.

The manuscript is well written and exposes results from the air monitoring campaign. In my opinion, the manuscript must be focused on that line of research and could be considered after major revisions.

Major comments:

1. The aim of the study is related to showing the results of the air monitoring campaign meanwhile the authors tried to include numerical simulation in their discussion-analysis with a small contribution to the analysis of the measurements during the campaign. The authors used a Gaussian Model without considering the photochemical reactions of PAHs in the air, even when they mentioned the formation of the OPAHS from the photochemical degradation of PAHs (Line26 p2). In my opinion, the authors must consider a more confident model (like Eulerians) to replicate the observations.

Response: The photochemical degradation of particle phase PAH is a very slow process – for most particle bound compounds it takes several hours or even days (Keyte et al., 2013*). During winter at high latitudes, with relatively little solar radiation and low temperatures, photochemical oxidation processes will be less important. This is true also for the formation of OPAHs from their parent-PAHs. Since transport time from local emissions to the sampling sites is very short (minutes) in comparison to photochemistry we consider the particle bound PAH to be inert, which justifies using a Gaussian model without photochemistry. This means that we can safely assume that the OPAH concentrations are from direct emissions rather than photochemical formation of locally emitted PAH. Some OPAHs are long-range transported to our site, and they may have been formed in photochemical reactions, but we only include local emissions in the dispersion modelling – long-range transport is added to the modelled concentrations based on background measurements.

We also want to stress that the Gaussian air quality dispersion model used here has been described and used successfully in several peer reviewed studies, both epidemiological and health impact assessments (examples listed below). Both in this study and in many other, it is not the dispersion model but the emission input that are most critical when it comes to uncertainties in the modelling. Especially in the case of wood burning (see also below).

**Keyte, I.J., Harrison, R.M. and Lammel, G: Chemical reactivity and long-range transport potential of polycyclic aromatic hydrocarbons – a review, Chem. Soc. Rev., 42, 9333 - 9391, https://doi: 10.1039/c3cs60147a, 2013.*

Examples of publications using the Gaussian AQ dispersion over Greater Stockholm:

Dreij, K., et al. Cancer Risk Assessment of Airborne PAHs Based on in Vitro Mixture Potency Factors. Environmental Science & Technology 2017 51 (15), 8805-8814. DOI: 10.1021/acs.est.7b02963.

Johansson, C.; Burman, L.; Forsberg, B. The effects of congestions tax on air quality and health. Atmos. Environ. 2009, 43, 4843–4854.

Kriit, H., Nilsson Sommar, J., Forsberg, B., Åström, S., Svensson, M., Johansson, C. A health economic assessment of air pollution effects under climate neutral vehicle fleet scenarios in Stockholm, Sweden. Journal of Transport & Health, 22, 101084, 2021.

Ljungman, P.L.S.; Andersson, N.; Stockfelt, L.; Andersson, E.M.; Nilsson Sommar, J.; Eneroth, K.; Gidhagen, L.; Johansson, C.; Lager, A.; Leander, K.; Molnar, P.; Pedersen, N.L.; Rizzuto, D.; Rosengren, A.; Segersson, D.; Wennberg, P.; Barregard, L.; Forsberg, B.; Sallsten, G.; Bellander, T.; Pershagen, G. Long-term exposure to particulate air pollution, black carbon and their source components in relation to ischemic heart disease and stroke, Environ. Health Perspect. 127 (10), 2019. https://ehp.niehs.nih.gov/doi/10.1289/EHP4757

Segersson, D.; Eneroth, K.; Gidhagen, L.; Johansson, C.; Omstedt, G.; Nylén A.E.; Forsberg, B. 2017. Health Impact of PM10, PM2.5 and Black Carbon Exposure Due to Different Source Sectors in Stockholm, Gothenburg and Umea, Sweden. Int J Environ Res Public Health | 14 (7), 2017. https://www.mdpi.com/1660-4601/14/7/742

Segersson, D.; Johansson, C.; Forsberg, B. 2021. Near-Source Risk Functions for Particulate Matter Are Critical When Assessing the Health Benefits of Local Abatement Strategies. Int J Environ Res Public Health, 18, 2021. https://www.mdpi.com/1660-4601/18/13/6847

Sommar et al., 2021. Long-term exposure to particulate air pollution and black carbon in relation to natural and cause-specific mortality: a multicohort study in Sweden. BMJ open, 11, 2021. 10.1136/bmjopen-2020-046040. https://bmjopen.bmj.com/content/11/9/e046040

Wu et al., 2022. Air pollution as a risk factor for Cognitive Impairment no Dementia (CIND) and its progression to dementia: A longitudinal study. Environment International, 160, February 2022, 107067.

In Section “2.4.4 Model simulations”, the text will be revised to justify the use of Gaussian model as below.

“All dispersion modelling of local emissions was performed with a Gaussian plume model in the Airviro air quality management system (Apertum, 2021; Segersson et al., 2017). <u>The photochemical degradation of particle phase PAH is a very slow process (Keyte et al., 2013) and the local emission source was very close to the sampling sites in the study. Therefore, a Gaussian model without photochemistry can be used, assuming that the OPAH concentrations are from direct emissions rather than photochemical formation of locally emitted PAH.</u>*

2. P7 L15 Could be more detailed about how the climatology data was used and applied in the study (ie: the data was averaged? are measured data in specific places? how many places?) How the historical data could fit the simulation in the period of analysis? The authors must justify this method to reduce the uncertainties about meteorological conditions for the PM dispersion.

Response: We use detailed data from a meteorological mast, in addition to the regular data (wind speed, wind direction, temperature, relative humidity, solar radiation), we have standard deviation of vertical wind speed and differential temperature to estimate the stability and mixed layer height. Hourly meteorological data was used to model the local contribution of B[a]P at the measurement sites. Using modeled local contributions with added measured monthly regional background from the same month we derived total B[a]P concentrations that was compared to measured values in Stockholm County. However, the climatology was used to model local B[a]P in the entire Stockholm County and it should also be noted that we added the average regional background during 2014-2018 to obtain total concentrations. This procedure was chosen in order to assess a general exposure and health impact in which we do not want to introduce a bias due to the conditions during our measurement campaign.

The climatology consists of frequency of occurrence of hourly mean data in 60 wind sectors, each 6^{o}, with 6 stability classes. All events falling into a specific sector will be classified according to the atmospheric stability conditions (as default discriminated by 6 intervals of Monin-Obukhov lengths). When all data have been sorted, frequencies of all classes will be estimated and the median values (of the Monin-Obukhov lengths) of each class (in this case 360 classes) are identified, including the specific date/hour when each class example occurred.

In order to extrapolate over the whole model domain, we use the diagnostic wind model that takes into account topography, roughness and land-use.

In Segersson et al (2017) we demonstrate the consistency in averages estimated using the two methods by comparing with hourly mean time series calculations for 7000 receptor points in the same domain as in this study. The comparison shows that the results are very similar; slope = 0.891 ± 0.002; intercept = 0.071 ± 0.021; correlation, r = 0.99, indicating that the climatology based calculations well represents the full hourly time-series.

In Section "2.4.4 Model simulations", the text will be revised accordingly:

"Airviro uses a diagnostic wind model based on the theory first described by Danard (Danard, 1977), in which it is assumed that small scale winds can be seen as a local adaptation of large-scale winds. Calculation of B[a]P concentrations was performed for local wood and traffic sources in a 5.5 km^2 size area with a 100 m grid size around the measurement sites using meteorology during the measurement period from a 50 m meteorological mast located in Högdalen, a suburb in Stockholm. <u>Meteorological data includes the wind speed, wind direction, temperature, relative humidity, and solar radiation.</u> Modelled monthly mean concentrations due to local residential wood combustion and road traffic at the Stockholm county measurement sites was based on hourly time series calculations using hourly meteorological from the Högdalen mast. The measured monthly concentration of B[a]P in regional background was added to the calculated local amount and the total B[a]P concentration can then be compared with the measured concentration during the measurement campaign in Stockholm urban background (TK), residential area (EN) and (YJ). The measurement in the northern residential area (DE) is outside of the emission inventory area and cannot be compared with calculated concentrations.

Dispersion modelling was also done for entire Stockholm County for local wood and local traffic sources using climatology for 1993-2010. <u>The climatology data consists of frequency of occurrence of hourly mean data in 60 wind sectors, each 6^o, with 6 stability classes, which are classified according to the atmospheric stability conditions (as default discriminated by 6 intervals of Monin-Obukhov lengths). After data sorting, frequencies of all classes is estimated and the median values (of the Monin-Obukhov lengths) of each class (in this case 360 classes) are identified, including the specific date/hour when each class example occurred. We use the diagnostic wind model that takes into account topography, roughness and land-use in order to extrapolate over the whole model domain.</u> The climatology derived modelled data was used to calculate yearly B[a]P concentrations in entire Stockholm County in 100x100 m grid size in order to obtain a general exposure and health impact assessment for average meteorological conditions. The average regional background B[a]P concentrations during the years 2014-2018 was added to the modelled local contribution. "

3. P13 L15 The authors showed the causes for the poor correlation between modeled and observed values of PAHs. This result does not contribute to the analysis. The numerical simulation could probably be erased from the manuscript due to the several causes mentioned by the authors and the limitation of the model used.

Response: We appreciate the referee's comment, but as we point out that the model calculations of concentrations due to emissions from residential wood combustion is very uncertain due to the many uncontrollable factors associated with wood burning, like wood characteristics (wetness, type of wood…), burning efficiency that depends not only on types of stoves but also on individual wood users' behavior etc. Even if there are these uncertainties, dispersion modelling is necessary in order to estimate spatial variations and population exposures, which we believe adds important information to the paper (also pointed out by referee no 1.).

Minor comments:

4. p4 subsection 2.2 Why only used the PM10 filter and not PM2.5? The PM2.5 sampling filter could indicate more confidence in the long-range transportation and source contribution of the PAHs and OPAHs.

Response: We do not think using $PM_{2.5}$ would make it possible to distinguish long-range transport (versus locally emitted) PAH and OPAH since most (if not all) PAH and OPAH are in the submicrometer size fractions REFs, which means that the concentrations of PAH and OPAH will be the same in PM_{10} and $PM_{2.5}$. But the argument for using PM_{10} was that the current air quality legislation of PAHs is based on PM_{10} (EU 2005).

EU: Council Directive 2004/107/EC of the European Parliament and of the Council of 15 December 2004 relating to arsenic, cadmium, mercury, nickel and polycyclic aromatic hydrocarbons in ambient air [2005] OJ L23/3.

5. p5 Title subsection 2.3.3 Change Levoglucosna by Levoglucosan

Response: The type will be corrected to “2.3.3 Levoglucosan, mannosan and galactosan”.