We show our gratitude to Anonymous Referee #1 for his constructive comments. We have revised the manuscript accordingly. Please find our point-to-point responses below.

Response to Anonymous Referee #1's comments

The authors observed Lung Deposited Surface Area (LDSA), which is an indicator of the adverse effects of nanoparticles on human health, in urban sites and their backgrounds, and explained their behavior and characteristics along with other parameters. As an important works of the authors, furthermore, they demonstrated to better estimate the LDSA concentrations from several widely monitored atmospheric and meteorological parameters, and characterize the statistical relationship with other parameters, by applied advanced statistical methods that combined automated input variables selection techniques with random effects. In the current that attracts public attention to the human effects of finer particles, new methods and results that better estimate this indicator, which may be better represented them than mass concentrations, from currently widely monitored parameters, have can be important implications to satisfy strong social demands in the near future.

The evaluation of the model and its usefulness are extremely and convincingly written in this manuscript. â□□However, I have some confusion concerns and questions regarding the interpretation of observed LDSA and some indicators analyzed by the authors. I hope the authors find my comments below useful. Therefore I would recommend the paper for publication after these clarifications.

Response: Thank you for the positive comments.

Specific comments:

Line 48-49: Clear information of particle size is misleading to the reader, because their information for particle deposition on the lungs has greater uncertainty by their various properties and also their mechanisms can be complex contribute. it necessary to clearly that these values are reference values.

Response: Thank you for your suggestion. We agree with you that the exact particle size can be inaccurate due to the reasons you mentioned. The idea of this paragraph is just to give a general information of how a pathway of a particle varies because of its particle size; therefore, we decided to remove the exact values, but to keep the terms 'coarser' and 'finer'.

Line 183-: How calibrations and corrections were made to compare the observations at several sites?

Response: The factory calibration of the instrument is based on reference number concentration aerosol, and response to LDSA is determined with 50 nm sized particles. Validation of the calibration was done by comparing the AQ Urban to other diffusion charging-based LDSA measurement devices. The instrument sensitivity with respect to LDSA is 0.215 μm^2 cm^{-3} fA^{-1} with the electrometer sensitivity being in the low fA range at 1 Hz operation. Better sensitivity can be achieved with longer integration times. Nominal integration time was 2 min but can be adjusted freely. According to the manufacturer, the internal precision of the AQ Urban is ± 3%, but this was not tested prior the campaign.

Table 1: Table 1: As the authors have descripted, previous studies of LDSA may have different target for deposition areas, which can lead to very different values. it should be clarified reviewed previous data from many sites.

Response: Thank you for the concern. Table 1 lists LDSA values from previous studies only at the alveolar region. This has been clarified in the caption of the table.

Line 330-: The contributed factors of the observed LDSA are discussed based on the time-series variations and the conclusions discussed in the previous studies observed at the same point. The characteristics of LDSA observed in this study should be more clearly based on the BC concentration data and analyzed backward trajectories, for example the effects of traffic and heating or the effects of long-distance transportation etc.

Response: Thank you for the suggestion. The observed LDSA concentrations were discussed based on Fig. 4 where we presented the diurnal cycle of LDSA. We also suggested in the original texts the possible underlying reasons for the variation, including long-range transport, human activities from traffic and heating. We improved the texts by including also the monthly frequencies of backward trajectory (Fig. S5). It shows that pollutants can be originated 600 km away from Helsinki in the winter by horizontal dispersion. Moreover, by showing the ratio LDSA/BC (Fig. S7 and S8), we also deduced that vehicular combustion emits high concentration of BC compared to its contribution to LDSA concentration.

Figure 4: Sufficient evidence is needed to explain that the value increased from the background is LDSA caused only by particles emitted in urban. In particular, it is unlikely that particles observed in background site will be observed in urban site as well without increase or decrease. Do previous studies, in particular, fully explain the long-term observations in this study?

Response: The diurnal and weekly variations in air pollutants were not clear as there were no anthropogenic activities nearby. Diurnal variation was mainly caused by the variation in the convective boundary layer height that caused mixing and dilution and not by local anthropogenic sources, which is expected for a regional background station (Luoma et al., 2021). We here made an assumption that the concentration we measured in regional background site was mostly background concentration influenced by local meteorology. The LDSA concentration at DH the after being subtracted from RB can then be regarded as the local increments to help identify the source apportionment. Similar calculation of simple subtraction was also done for example in Kuula et al. (2020) and in Jafar and Harrison (2021). To make it clear in the text, we re-organised the paragraph with one more citation.

Line 368-: The importance and implications of the ratio of LDSA to some of the parameters shown here are need to more clear.

Response: Thank you for the suggestion. The main idea of presenting the ratio of LDSA to BC, NOx, PNC and PM2.5 is to demonstrate the contribution of LDSA. For example, at the SC site, the relatively high LDSA/PM2.5 during morning peak hour reveals that the vehicular combustion emits smaller particles, which elevate the LDSA concentration but meanwhile do not substantially influence the value of PM2.5. On the other hand, the low value of LDSA/BC, LDSA/NOx and LDSA/PNC during the same peak period show the opposite. This can be explained by the fact that vehicular combustion emitted high concentration of BC, PNC and NOx compared to its contribution to LDSA concentration.

The calculation of the ratio is another way to validate the fact that urban activities, such as vehicular combustion, have different impacts on different air quality parameters. I understand that expressing the relationship in ratio is not the most straight-forward way to show the idea, but I believe it shows the contribution of LDSA from another perspective.

In particular, LDSA, which is measured by diffusion charge, is based on the relationship that the amount of charge measured is proportional to about 1.1 power of the particle size. On the other hand, PM2.5 and PINC are proportional to the 1st or 2nd power of the particle size, so in theory these three parameters are explained only by the different dimensions for diameter.

Response: Thank you for the concern. LDSA by Pegasor effectively measures the deposited surface area of ~0.01–0.4 µm and you are right that it should be proportional to about 1.1 power of the particle size after considering the deposition factor. PM2.5, however, measures mass concentration of particles smaller than 2.5 µm. PM2.5 should be proportional to the 3rd power of the particle size in the measuring range because it depends on the volume and the density of the particles. PNC in this study measures number concentration of particle sized between 0.03 to 1 µm and it should not be a function of particle size. Due to the different measuring ranges and the nature of the three parameters, I believe that these three parameters could not be explained only by the different particle sizes.

Moreover, the trend of diurnal variation in the ratio of LDSA to some components seems to be inconsistent with the relationship with the factors of LDSA explained in the previous section. This reason seems to be due to the fact that, for example, BC is a mass-based concentration, whereas LDSA is different, as mentioned earlier. Therefore, LDSA may potentially not have a linear relationship with PM2.5 and BC concentrations in theoretical, but does it affect the model constructed by these complex regressions?

Response: Thank you for raising this concern. You are right that PNC, LDSA and PM2.5 that respectively represent the number, surface and mass concentration do not have similar distribution and they do not necessarily have linear relationship. In order not to violate the assumption of the regression model, we converted the distribution of aerosol and trace gases into normal distribution by performing logarithm transformation. Also, we conducted statistical tests of the residuals of the regression model to check if they satisfy the requirements for the regression model.

Line 418-: The estimation results that showed different performance depending on the variables selected were clearly explained. Is it possible to quantitatively discuss the contribution of the parameters involved in LDSA, especially with some of the coefficients shown in Table 5?

Response: Thank you for the suggestion. Apart from counting the times the involved parameters appeared in the LDSA estimation, it is also good to refer to the Pearson correlation coefficient (r) of LDSA with those involved parameters in order to show their contribution to the LDSA estimation. Technically, the r values can effectively reflect the overall contribution. Therefore, we decided to refer some of the r values when we discuss the relative contribution in LDSA estimation (See Ln 413 and Ln 421–424). However, the coefficients shown in Table 4 and 5 could not tell the relative contribution because the ranges and magnitudes of those involved parameters are different. They, on the other hand, give the exact coefficients to calculate the output of LDSA.

Reference:

Jafar, H. A., and Harrison, R. M.: Spatial and temporal trends in carbonaceous aerosols in the United Kingdom, Atmos. Pollut. Res., 12, 295-305, https://doi.org/10.1016/j.apr.2020.09.009, 2021.

Kuula, J., Kuuluvainen, H., Niemi, J. V., Saukko, E., Portin, H., Kousa, A., Aurela, M., Rönkkö, T., and Timonen, H.: Long-term sensor measurements of lung deposited surface area of particulate matter emitted from local vehicular and residential wood combustion sources, Aerosol Sci. Tech., 54, 190-202, https://doi.org/10.1080/02786826.2019.1668909, 2020.

Luoma, K., Niemi, J. V., Aurela, M., Fung, P. L., Helin, A., Hussein, T., Kangas, L., Kousa, A., Rönkkö, T., Timonen, H., Virkkula, A., and Petäjä, T.: Spatiotemporal variation and trends in equivalent black carbon in the Helsinki metropolitan area in Finland, Atmos. Chem. Phys., 21, 1173-1189, https://doi.org/10.5194/acp-21-1173-2021, 2021.