Dear referee,

Thank you for your comments and suggestions. We carefully addressed them one by one as shown below. Hope you find our revisions useful. Thank you again.

Regards,

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#Referee 1

The authors present results from a land use regression (LUR) framework used to create 500m resolution exposure fields for multiple pollutants, including NO2, O3, total PM1- and PM2.5, and multiple PM10 species. Their LUR models have good predictive performance in leave one out cross validation. I agree with the authors that the fine-scale exposure products may be useful for future exposure and epidemiological studies. I believe, however, that the study does not go far enough in explaining the implications of their results. In particular, I find it is lacking in two areas relevant to the scope of ACP.

First, very little description is given of the meaning behind the variables that are selected for the LUR models, including why they are predictive of the various species and what they may be proxies for. Did any variables that have been found predictive not make sense? Are there co-linearities that may be obscuring the influence of some variables over others? Can we learn something from the predictors chosen that can inform policies to reduce exposure?

Response: The manuscript has provided descriptions to explain the associations between air pollutants and their predictive variables. The descriptions can be found in Line 221-226, Line 231-247, Line 150-151 and Line 273-280. The explanations were confirmed by the supports of previous research. Thus, the meaning behind the relationships were clearly demonstrated.

It is true that some variables may have a certain level of co-linearities. Similar to previous studies, we applied Variance Inflation Factor (VIF) measures the severity of multicollinearity in regression analysis. Predictor variables with a value higher than 5.0 were removed (de Hoogh et al., 2013; Gulliver et al., 2018; Hsu et al., 2018; Jones et al., 2020; Larkin et al., 2017; Ma et al., 2019; Zhang et al., 2015). So, it is believed that co-linearities should not induce major issues.

The model was demonstrated its capabilities to simulate the multiple air pollutants, and to reflect the relationships between each predictive variable and various air pollutants. Similar to other studies (Vizcaino and Lavalle, 2018; Shi et al., 2020), our model can be used to evaluate any potential air pollutant emission control policies especially for those targeting to multiple air pollutants.

The descriptions in the main paper:
Line 221-226: The predictor variables included in the O_3 LUR model were the number of total vehicles in a buffer size of 700 m, longitude, and the area of urban green space within a 300-m buffer. The predictor variable of vehicles had a negative effect on O_3 concentration. This negative effect reflects the titration of O_3 in urban areas with a large amount of NO and NO_2 emitted by traffic. Longitude has a positive effect on O_3 concentration, suggesting the influence of regional transported air masses. A LUR study in Nanjing, China also included longitude in the final O_3 model (Huang et al., 2017). Urban green space had a positive effect on O_3 concentration, which is probably due to biogenic volatile organic compounds as the precursors of ozone formation (Ma et al., 2021; Ren et al., 2017).

Line 231-247: The PM_{10}, $PM_{2.5}$, and NO_2 LUR models included several predictor variables (e.g., the number of medium and heavy-duty vehicles in a buffer size of 500 m) representing vehicular emissions (Table S3). Thus, the concentration of PM_{10}, $PM_{2.5}$, and NO_2 is largely affected by the traffic emissions in Hong Kong, with higher concentration estimated along the road network. In addition, the relatively higher concentration of PM_{10}, $PM_{2.5}$, and NO_2 was estimated in areas with the high population density (e.g., the northern part of Hong Kong island, the Kowloon City district, and the Yau Tsim Mong district). PM_{10} and $PM_{2.5}$ had moderate positive corrections with NO_2, with Pearson correlation coefficient (PCC) values at 0.570 and 0.696, respectively (Table 1). Consistent with Li et al. (2022), the LUR model-derived concentration of PM_{10} TC, PM_{10} NO_3^-, and PM_{10} SO_4^{2-} was relatively higher at developed urban areas and along major roads. In contrast to this, the spatial distribution of PM_{10} Cd showed a north–south gradient, with relatively higher concentration in the northern part and relatively lower concentration in the southern part. These PM_{10} chemical species only had weak to moderate positive correlations with PM_{10} mass, with PCC values ranging from 0.189 to 0.589 (Table 1). For O_3, there was an increasing trend from west to east, suggesting the influence of transboundary pollution on the spatial distribution pattern. In addition, O_3 concentration was largely affected by traffic emissions, with lower concentration estimated along major roads compared with other areas. Due to nitric oxide titration (Han et al., 2023), O_3 concentration was generally negatively correlated by various degree with PM_{10}, PM_{10} chemical species, $PM_{2.5}$, and NO_2 (Table 1).

Line 150-151: Due to this procedure, the included predictor variables may obscure the potential influence of others.

Line 273-280: The results highlight that the synergistic control of multiple emission sources and key precursors is urgently needed for the joint control of multiple air pollutants (Saha et al., 2020; Yim et al., 2019). For instance, the Hong Kong government has spent tremendous efforts on the reduction of vehicular emissions over the past two decades, which successfully reduced traffic-related air pollutants like $PM_{2.5}$ and NO_2. However, as revealed by the present study and previous studies (HKEPD, 2022; Zeng et al., 2022), O_3 pollution has become an emerging issue, especially in rural areas of Hong Kong, which cannot be accomplished *through the control of vehicular emissions. Thus, more research efforts should be conducted to understand the complex and varying interaction of emission sources, pollutant sensitivity to precursors, and air quality in a city to formulate more effective and specific air quality management policies.*

Second, there is little description provided of the exposure products themselves. What are the implications for human exposure? It would be useful to select important areas of the city (e.g., an area with high population density), describe which pollutants are predicted to have high concentrations, and offer some suggestions about why.

Response: This manuscript mainly focused on the development of an integrated model framework. Due to the length limit, it is not feasible to provide detailed description of the exposure products. Nevertheless, we did try our best to describe the exposure results. Section 3.3 provided spatial distribution map in section 3.3 , while the description in Line 267-280 highlighted the mechanisms and insights for pollution control.

The descriptions in the main paper:

3.3 Spatial distribution maps: The spatial distribution maps of multiple air pollutants derived from established LUR models are shown in Figure 2, whereas Table S4 shows the statistical description of the estimated air pollutant concentration. The PM_{10}, $PM_{2.5}$, and NO_2 LUR…

Line 267-280: The spatial variation of air pollution can be used for hotspot identification for air quality management and exposure assessment in epidemiological studies using the geospatial locations of the subjects (Crouse et al., 2015; Jones et al., 2020; Li et al., 2021). The major explanation for the spatial differences in concentration of multiple air pollutants was the differences in their emission sources (Cai et al., 2020; Jin et al., 2019; Levy et al., 2014; Wu et al., 2017). For instance, $PM_{2.5}$ and NO_2 are more linked to traffic and industrial emissions in developed urban areas, while relatively high O_3 concentration in rural areas is formed through complex chemical reactions between biogenic volatile organic compounds and nitrogen oxides (Table S3 and Figure 2). The results highlight that the synergistic control of multiple emission sources and key precursors is urgently needed for the joint control of multiple air pollutants (Saha et al., 2020; Yim et al., 2019). For instance, the Hong Kong government has spent tremendous efforts on the reduction of vehicular emissions over the past two decades, which successfully reduced traffic-related air pollutants like $PM_{2.5}$ and NO_2. However, as revealed by the present study and previous studies (HKEPD, 2022; Zeng et al., 2022), O_3 pollution has become an emerging issue, especially in rural areas of Hong Kong, which cannot be accomplished through the control of vehicular emissions. Thus, more research efforts should be conducted to understand the complex and varying interaction of emission sources, pollutant sensitivity to precursors, and air quality in a city to formulate more effective and specific air quality management policies.

Specific comments

Abstract: it is important to describe the exposure product in full, including the temporal coverage (i.e., which year?)

Response: Revised as suggested (Line 20-21).
Line 20-21: …2017 annual-average exposures of four major PM_{10} chemical species as well as four criteria air pollutants of PM_{10}, $PM_{2.5}$, NO_2, and O_3 in…

Line 81-82: it strikes me as strange to have 3 citations for a sentence describing the geography of Hong Kong.

Response: Revised with only one reference kept (Line 85).
Line 85: …the southeast coast of the Pearl River Delta (PRD) region, China (Yim et al., 2009).

122-123: it would be helpful to explain more about why lat/lon would account appropriately for transboundary pollution

Response: We added the capacity of using lat/lon as predictor variables in the revised manuscript (Line 128-129).

Line 128-129: …, the geo-locations of longitude and latitude were also adopted which could reveal a north-south or west-east gradient of air pollutant concentrations (Huang et al., 2017).

134-135: please explain "if the direction was as pre-defined"

Response: We revised the text to improve this description (Line 142).

Line 142: …the direction was consistent with the pre-defined one.

135: model selection process: please confirm whether the model selection R2 was calculated on the training dataset or the hold out dataset. If the training dataset, is there risk of over-fitting?

Response: The calculation was on the full dataset (Line 140). We then further used leave-one-out cross-validation (LOOCV) method to validate the established model (Line 158).

Line 140: Using the full dataset, we ranked…
Line 158: Leave-one-out cross-validation (LOOCV) was used…

135: It seems to me that more flexible machine learning methods may be more adept at capturing nonlinear relationships between environmental predictors and measured pollutants. Is it expected that these variables have a linear relationship with pollutant species? The final paragraph of the manuscript mentions that these are more appropriate with more data, but there is no evidence given or description of how much data is needed.

Response: It is correct that environmental predictors and measured air pollutants may have nonlinear relationships. Nevertheless, LUR models are still the widely used model for epidemiological research because they are well proved to reflect the relationships between predictive variables and targeted air pollutants. There are still a large number of air quality modelling studies using LUR models (de Hoogh et al., 2013; Gulliver et al., 2018; Hsu et al., 2018; Jones et al., 2020; Larkin et al., 2017; Ma et al., 2019; Zhang et al., 2015; Vizcaino and Lavalle, 2018; Shi et al., 2020). We agree that machine learning method is another a useful tool for air quality modelling, so our main paper has included the further research perspective on using various machine learning algorithms (Line 305-306).

Regarding the required data set for LUR/machine learning model development, it is an important research topic but out of the scope of the present study, which mainly proposes an integrated model framework for accurate multi-air-pollutant exposure assessments in high-density and high-rise cities.

Line 305-306: Apart from the LUR approach, other spatiotemporal statistical modelling methods, such as various machine learning algorithms, should be applied when a larger data set is available.

167: "either comparable to or higher than" makes it sound like a dichotomous variable. I think "generally greater than" would describe this well enough.

Response: Revised as suggested (Line 182).

Line 182: ... generally higher than…

176-178: I do not understand how the selection of the predictor variables was related to these other factors. Please clarify.

Response: We revised the mentioned content to make it clearer (Line 191-192).

Line 191-192: The selection of these predictor variables was driven by the emission sources…

192: This line refers to a negative z score close to 1. Does this suggest anti-spatial correlation for Cd? More description of this variable is needed

Response: We added the description of Moran's *I* and the corresponding *z*-score and *p*-values in the revised manuscript (Line 152-156).

Line 152-156: The spatial autocorrelation (Moran's I) tool measures spatial autocorrelation using both feature locations and feature values simultaneously. Meanwhile, z-score and P-values were calculated to evaluate the significance of Moran's I value. z-score values are standard deviations, whereas the Moran's I index is bounded by -1.0 and 1.0. When the z-score or P-value indicates statistical significance, a positive Moran's I index value indicates tendency towards clustering, whereas a negative Moran's I index value indicates tendency towards dispersion (Cordioli et al., 2017; Luminati et al., 2021). Moran's I index and the corresponding z-score and P values on…

Could the authors provide spatial maps of error at monitor locations? In general, it would be useful to develop estimates of uncertainty on the the same spatial scale as the predictions.

Response: We have added the spatial maps of prediction error [differences between predictions and observations] (Figure S7 in SI) and error fraction [(predictions-observations)/observations] (Figure 2) of the LUR models to show the uncertainty/error. The related content is added (Line 186-189).

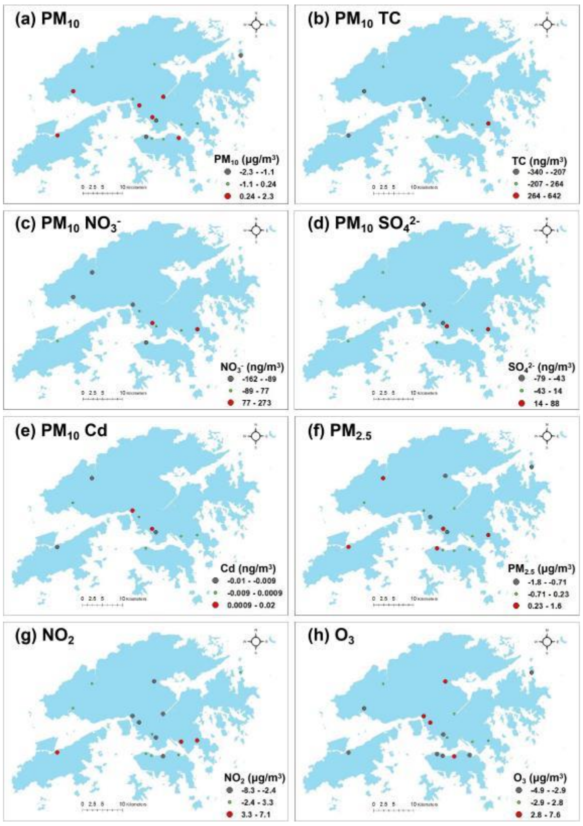

Figure S8. The distribution of prediction errors [predicted concentration – observed concentration] of the established LUR models.

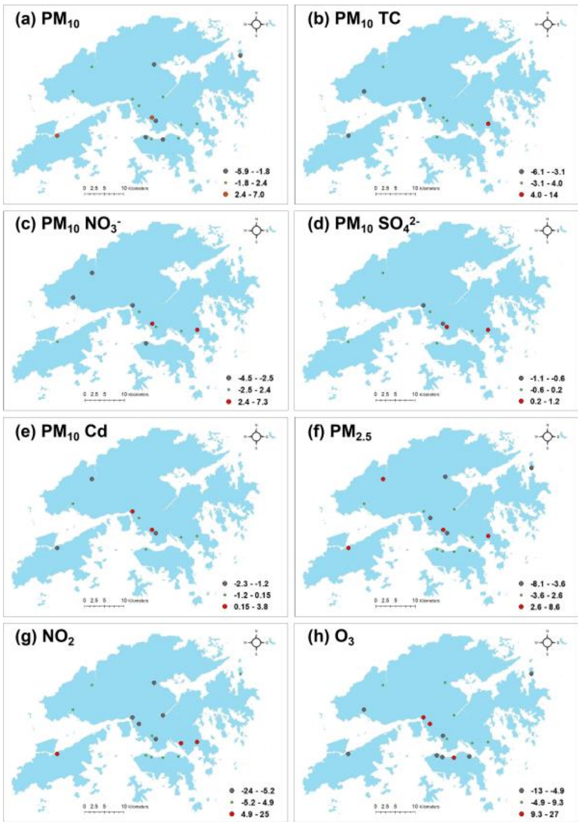

Figure 2. The distribution of prediction error fractions (%) of the established LUR models. The prediction error fraction is defined as [(predicted concentration – observed concentration)/observed concentration].

Line 186-189: The prediction error fractions of the LUR models ranged between -5.9%–7.0%, -6.1%–14%, -4.5%–7.3%, -1.1%–1.2%, -2.3%–3.8%, -8.1%–8.6%, -24%–25%, and -13%–27%, respectively, for PM_{10}, PM_{10} TC, PM_{10} NO_3^-, PM_{10} SO_4^{2-}, PM_{10} Cd, $PM_{2.5}$, NO_2, and O_3 (Figure 2).

220: is this implying that population density leads to high PM? Can the authors suggest a mechanism here that isn't explained by the other model covariates?

Response: Population density can serve as a proxy to reflect the contribution of other emission sources due to human activities (i.e. cooking and industrial emissions) that were not included in the model. We do include the explanation on this mechanism (Line 269-271).

Line 269-271: The major explanation for the spatial differences in concentration of multiple air pollutants was the differences in their emission sources (Cai et al., 2020; Jin et al., 2019; Levy et al., 2014; Wu et al., 2017). For instance, $PM_{2.5}$ and NO_2 are more linked to traffic and industrial emissions in developed urban areas, …

239-241: I am confused about the difference between the GAS and PM modules. Is it necessary to differentiate beyond the pollutant species names?

Response: In the revised manuscript, we added the reason for separating PM and GAS modules (Line 76-79).

Line 76-79: The PM and GAS modules were separated because the measurement and LUR modelling of PM species and gaseous pollutants are largely different in terms of measurement techniques, the number of required measurement sites, and selected predictor variables, etc. the In the present study, the…

257-260: I am not sure what this adds to the discussion. Can the authors be more specific here based on the predictors chosen for the final model?

Response: This part aimed at providing a description to link our established models to policy evaluation. We have revised the mentioned content to make it more specific (Line 274-280).

Line 274-280: For instance, the Hong Kong government has spent tremendous efforts on the reduction of vehicular emissions over the past two decades, which successfully reduced traffic-related air pollutants like $PM_{2.5}$ and NO_2. However, as revealed by the present study and previous studies (HKEPD, 2022; Zeng et al., 2022), O_3 pollution has become an emerging issue, especially in rural areas of Hong Kong, which cannot be accomplished through the control of vehicular emissions. Thus, more research efforts should be conducted to understand the complex and varying interaction of emission sources, pollutant sensitivity to precursors, and air quality in a city to formulate more effective and specific air quality management policies.

How was 500m selected as the best resolution for the predictions?

Response: Based on the spatial resolution of the predictor variables. We revised the related content to make it clearer (Line 169-170).

Line 169-170: A spatial resolution of 500 m ×500 m was adopted here due to the spatial resolution of most predictor variables is at several hundred meters resolution.

Reference:

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