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Development and validation of land use regression models for ultrafine particles in Augsburg and Regensburg, Germany

Marco Dallavalle [a,b], Josef Cyrys [b], Susanne Sues [c], Simonas Kecorius [b], Susanne Breitner-Busch [a,b], Regina Pickford [b], Alexandra Schneider [b], Annette Peters [a,b], Kathrin Wolf [b,*]

[a] Chair of Epidemiology, IBE, Faculty of Medicine, LMU Munich, Germany
[b] Helmholtz Zentrum München, German Research Center for Environmental Health (GmbH), Institute of Epidemiology, Neuherberg, Germany
[c] Environmental Science Center, University of Augsburg, Augsburg, Germany

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ABSTRACT

Ultrafine particles (UFP) are suspected to have a high toxic potential, but evidence from long-term epidemiological studies remains sparse since highly spatially resolved UFP data is lacking. We modelled long-term annual average total particle number concentration (PNC) as indicator for UFP for two middle-sized German cities (Augsburg and Regensburg) and their surroundings, which are part of the German National Cohort (NAKO), for subsequent linkage with health data.

Supervised land use regression (LUR) models were developed for Augsburg, combining two previous measurement campaigns (monitoring sites: 2014/15: $N = 20$ and 2017: $N = 6$) and spatial predictors. To account for the time difference and repeated monitoring sites, we applied a generalized additive model (GAM) and a mixed model (MM). Models were internally validated using leave-one-out cross-validation (LOOCV). We transferred the models to the Regensburg region and externally validated our predictions using in-situ measurements carried out in 2020/21 at six monitoring sites.

For both approaches, models showed highly adjusted explained variance and LOOCV R^2 (GAM: 0.90 and 0.76; MM: 0.91 and 0.86). Similar predictors were selected, mainly indicators for road network and industrial areas. The external validation showed good agreement of measured and predicted PNC with Spearman correlation coefficient $r = 0.75$ (GAM) and 0.86 (MM), though both models tended to underestimate the concentrations.

The two LUR models resulted in similar predictions and captured intra-city spatial patterns and city-rural gradients well. The Augsburg models could be effectively transferred to Regensburg since the study regions featured similar characteristics. To evaluate the predictive capability in novel study areas, external validation measurements are recommended.

1. Introduction

Ultrafine particles (UFPs, particles with a diameter < 100 nm) are suspected to have a high toxic potential due to their small particle

* Corresponding author at: Institute of Epidemiology – Research Group “Environmental Risks”, Ingolstädter Landstr. 1, 85764 Neuherberg, Germany.

E-mail address: kathrin.wolf@helmholtz-munich.de (K. Wolf).

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size, large surface area and high surface reactivity (HEI Review Panel on Ultrafine Particles, 2013; Peters et al., 2011). However, evidence from epidemiological studies remains sparse mainly due to the lack of routine monitoring that characterises variability of UFPs in space and time (Morawska et al., 2019; Ohlwein et al., 2019; Vallabani et al., 2023). This lack of data hinders the development of suitable prediction models for accurate exposure assessment. In 2021, the World Health Organization (WHO) updated the air quality guidelines for particulate matter <2.5 μm ($PM_{2.5}$) and other air pollutants based on new scientific findings (World Health Organization (WHO), 2021). A good practice statement of the WHO also recommends integrating UFP monitoring (measured as total particle number concentrations (PNC)) into existing air quality monitoring and advancing exposure assessment for epidemiological studies. UFP data capturing the spatial variability are particularly needed to improve our understanding of their long-term health effects.

LUR models have been widely used for traditional and criteria air pollutants like $PM_{2.5}$ or NO_2 but only few studies modelled UFP or PNC (Eeftens et al., 2016; Hoek, 2017; Hoek et al., 2011; Ma et al., 2024). PNCs vary on a smaller scale than particle mass concentrations and previous efforts mostly build on mobile monitoring which capture PNC by local sources but not adequately characterize annual average surfaces. We previously developed a supervised land use regression (LUR) model for PNC for the Augsburg region, Germany (Wolf et al., 2017) which we linked to the population-based Cooperative Health Research in the Region of Augsburg (KORA) cohort and observed adverse effects on inflammatory markers (Pilz et al., 2018; Vogli et al., 2023), insulin sensitivity (Zhang et al., 2021), metabolic alterations (Yao et al., 2022), incident distal sensorimotor polyneuropathy (Herder et al., 2023), cardiometabolic phenotypes (Woeckel et al., 2024), prevalent diabetes and obesity (Niedermayer et al., 2024). Since then, additional PNC measurements have been conducted in the Augsburg region in 2017 (Giemsa et al., 2021) and the amount of spatial predictors has been increased with remote sensing. Also, the German National Cohort (NAKO), the largest population-based cohort in Germany (Peters et al., 2022) including a deep characterization of individual-level risk factors and health variables was incepted in 2014 and offers a huge potential for environmental health analyses. This study aimed to develop robust LUR models for long-term annual average PNC enabling accurate exposure assessment for a large epidemiological study, to assess how well the models can be transferred between specific regions, and to identify key local factors that need to be considered in the process. Therefore, we modelled PNC as an indicator for UFP for two NAKO study regions (Augsburg and Regensburg) for subsequent linkage with health data. Specific objectives were to i) improve our LUR model for Augsburg by using additional measurements, an extended set of spatial predictors and the addition of a temporal component to account for the time difference between the two measurement campaigns and ii) transfer the prediction model to Regensburg and validate the predictions with new PNC measurements from Regensburg. Our results will enhance the understanding of PNC dispersion in urban and rural areas, LUR model transferability between cities and ultimately, support evidence-based public health policies and urban planning.

2. Methods

2.1. Sampling campaigns in the Augsburg region

As part of the previous projects ULTRA3 (Environmental nanoparticles and health: Exposure, modelling and epidemiology of nanoparticles and their composition within KORA) and LfU (Bavarian Environment Agency), PNC measurements were conducted at 20 sites in Augsburg and two adjacent districts in 2014/15 (Wolf et al., 2017) and at six sites in Augsburg in 2017 (Giemsa et al., 2021) with four sites at the same location in 2014/15 and 2017 (Supplemental Fig. S1). We calibrated the discontinuous site measurements (ULTRA3: three measurement rounds of two weeks per site between 01.03.14–15.04.2015; LfU: eight measurement rounds of two weeks per site between 01.01.2017–31.12.2017) against the long-term annual average with continuous measurements from an urban background site (reference site) following our previous modelling approach (Wolf et al., 2017).

2.2. Spatial predictor data

We collected and harmonized spatial information (as close as possible to the years of the measurement campaigns), which has shown plausible associations with PNC but also other air pollutants in previous modelling efforts (Supplemental Table S1) (de Hoogh et al., 2018; Eeftens et al., 2016; Kerckhoffs et al., 2021; Wolf et al., 2017). Local land use, road network, topography, as well as a 3D building model were obtained from the German Federal Agency for Cartography and Geodesy. We also downloaded road network data from OpenStreetMap (https://www.openstreetmap.org/). Socioeconomic information, including population and household density, was acquired from a private company (WiGeoGIS GmbH). Meteorological predictors such as wind speed and precipitation height were downloaded from the Climate Data Center of the German Weather Service (https://opendata.dwd.de/). Satellite information on imperviousness and forest density was gathered from the European Environmental Agency. Additionally, we included the Normalized Difference Vegetation Index from Landsat8 and post-processed satellite data of tropospheric and near-surface nitrogen dioxide (NO_2) concentrations from Copernicus Sentinel and the German Aerospace Center. In total, we considered 198 candidate predictors which are summarized in Supplemental Table S1, together with their source, temporal coverage, buffer sizes, variable transformations, and anticipated direction of association with PNC.

2.3. LUR model building and internal validation

We mainly followed our previous supervised forward stepwise selection procedure and fit regression models with the long-term average concentration at the monitoring sites as the outcome and the potential predictor variables as explaining covariates (Wolf et al., 2017). To account for the time difference between the measurement campaigns (2014/15 vs 2017) and repeated monitoring sites

($N = 4$), we applied two well-established methods that offer a robust and flexible framework for analyzing repeated measurements: a generalized additive model (GAM) and a linear mixed effects model (MM). In the GAM, the measurement year was incorporated as a factor variable, and the centred X/Y coordinates as a bivariate smooth function (regression spline with three degrees of freedom) to model potential non-linear relationships. The MM included the sites as random effects to account for their individual variability. Before the actual predictor selection, we pre-selected predictors by excluding those with extreme outliers (minimum or maximum value within the threefold of the 10th to 90th percentile range below or above the 10th and 90th percentile) or low variability (at least five sites exhibit differing values) to improve the stability of the models (Eeftens et al., 2016; Wolf et al., 2017). We then calculated a univariate regression for all potential predictors and selected the predictor with the highest R^2. Further predictors were then selected step by step by maximizing the model-adjusted explained variance R^2 (if the increase was >1 %, the direction of the effect was as expected, and there was no change in the direction of other predictor estimates), limiting multicollinearity (variance inflation factor < 3) and excluding non-significant predictors ($p > 0.1$). The model performance was internally validated by leave-one-out cross-validation (LOOCV), thus the model selection process was repeated separately for each left-out monitor. To assess and visualize the predicted PNC for the entire Augsburg region, we calculated the selected predictors for the centroids of a 50 m*50 m grid and applied the final models to these centroids to compile grid maps.

2.4. Model transfer to Regensburg region and external validation

We directly transferred the final models from Augsburg to the Regensburg region, by calculating the selected predictors for the centroids of a 50 m*50 m grid, and applying the final models to these centroids. We externally validated our predictions by in-situ measurements at six monitoring sites (Supplemental Fig. S2) carried out in 2020/21 (Cyrys et al., *in preparation*). The monitoring sites included three urban background (sites 1–3), two urban traffic (4, 6), and one regional background site (5) to reflect the residential locations of the NAKO study participants. We measured PNC with three mobile condensation particle counters (Environmental Dust Monitor (EDM) 465, GRIMM Aerosol Technik, Germany) for six biweekly periods, accounting for seasonal variations. Before and after each individual measurement, a comparison measurement was carried out with all the instruments. Like the Augsburg measurements, we adjusted the discontinuous site measurements to the long-term annual average with continuous measurements from an urban background site (reference site, 7, Supplemental Fig. S2). We then calculated the Spearman correlation between the measured and predicted concentrations. Moreover, we visually evaluated the performance of model transferability by plotting the predicted PNC, the street network and the selected spatial predictors within 500 m around each site.

All analyses were conducted with QGIS version 2.6.1 (QGIS Association, 2020) and R version 4.3.1 (R Core Team, 2023).

3. Results

The measured PNC concentrations in the Augsburg region ranged from 5644 to 16,645 cm^{-3} with a median of 8393 cm^{-3}. The pre-selection of predictor variables led to the exclusion of 44 predictors, thus 154 candidate predictors were included in the stepwise selection. Both models performed well with an adjusted explained variance and LOOCV R^2 of 0.90 and 0.76 for the GAM for Augsburg and Regensburg and 0.91 and 0.86 for the MM, respectively (Table 1). Also, both models selected the same indicators for road network (inverse distance to major roads and bus stops and the road length of major roads in a 100 m buffer) and one predictor for industrial areas (GAM: 1000 m buffer; MM: 300 m buffer). The MM additionally selected near surface NO_2. The resulting grid maps for the Augsburg region showed similar spatial patterns, with the highest concentrations close to major roads (Fig. 1, top and Supplemental Fig. S3, top). However, the GAM featured higher contrasts with lower concentrations in the more rural areas. The spatial patterns for 2014/15 (Supplemental Fig. S3, top) and 2017 (Fig. 1, top) were generally very similar. However, annual means for 2017 were estimated considerably lower by the GAM with a range from 2267 to 21,672 cm^{-3} (median = 4639 cm^{-3}) compared to the MM predictions ranging from 3972 to 20,890 cm^{-3} (median = 5533 cm^{-3}) (Supplemental Fig. S4).

Table 1
Model specification, selected predictors and model performance measures of the Augsburg model.

	Model specification	Roads	Land use	Other predictors	Adj R^2	LOOCV Adj R^2
GAM	gam(PNC ~ as.factor(year) + s(X,Y,k = 4, fx = T) + predictors)	Inverse distance to nearest major road (BKG) Inverse logarithm of distance to nearest bus stop (OSM) Road length of major roads within 100 m (OSM)	Industry within 1000 m	–	**0.90**	**0.76**
MM	gamm4(PNC ~ predictors, random = ~(1| ID))	Inverse distance to nearest major road (BKG) Inverse logarithm of distance to nearest bus stop (OSM) Road length of major roads within 100 m (OSM)	Industry within 300 m	Near surface NO_2	**0.91**	**0.86**

GAM: Generalized additive model; MM: Linear mixed effects model; BKG: Federal Agency for Cartography and Geodesy; OSM: Open Street Map; Adj: adjusted; LOOCV: leave-one-out cross-validation.

For the Regensburg region, the transferred models showed similar patterns and contrasts (Fig. 1, bottom and Supplemental Fig. S3, bottom). The predicted PNC concentrations for 2017 ranged from 2516 to 21,494 cm^{-3} (median = 4526 cm^{-3}) for the GAM and from 3972 to 20,471 cm^{-3} (median = 5509 cm^{-3}) for the MM (Supplemental Fig. S4). Pearson correlation coefficient showed a high correlation with $r = 0.85$ for Augsburg and 0.83 for Regensburg confirming that the models captured similar patterns in PNC.

The external validation at the monitoring sites in Regensburg showed a very good agreement of measured and predicted PNC (Fig. 2) with a better performance of the MM (Spearman correlation coefficient $r = 0.86$ vs 0.75 for the GAM). Both models tended to underestimate the concentrations, especially at one traffic site (6), but the GAM also at two background sites (3 and 5). The visual comparison of the predicted PNC, the street network and the selected spatial predictors within 500 m around each site demonstrated the influence of the road network predictors, especially for the two urban traffic sites (4 and 6) but also two urban background sites (1 and 3; Supplemental Fig. S5). The predictors for industrial areas and near surface NO_2 showed less local influence due to their coarser spatial resolution.

4. Discussion

We refined our previously developed LUR model for PNC for the Augsburg region (Wolf et al., 2017) by using additional measurements, an extended set of spatial predictors and the addition of a temporal component to account for the time difference between the measurements. We then transferred the prediction model to the Regensburg region and validated our predictions with new PNC measurements. Since we were specifically interested to test the spatial transferability of the modelling approach, we did not pool the two regions together. We compared two different modelling approaches, which both performed well and resulted in similar predictions capturing intra-city spatial patterns and city-rural gradients. We found that the Augsburg model could be effectively transferred to Regensburg, as the geographic features and spatial variability of the predictors across the two study areas were comparable. Hence, we provide spatially highly resolved and standardized exposure maps for two NAKO study regions which will allow assessing the exposure to annual average PNC concentrations at the residential addresses of more than 30,000 NAKO participants (Peters et al., 2022). These maps will enable subsequent epidemiological analyses to investigate the long-term effects of PNC on various health outcomes adjusting for a wealth of individual characteristics but also further air pollutants and other environmental exposures.

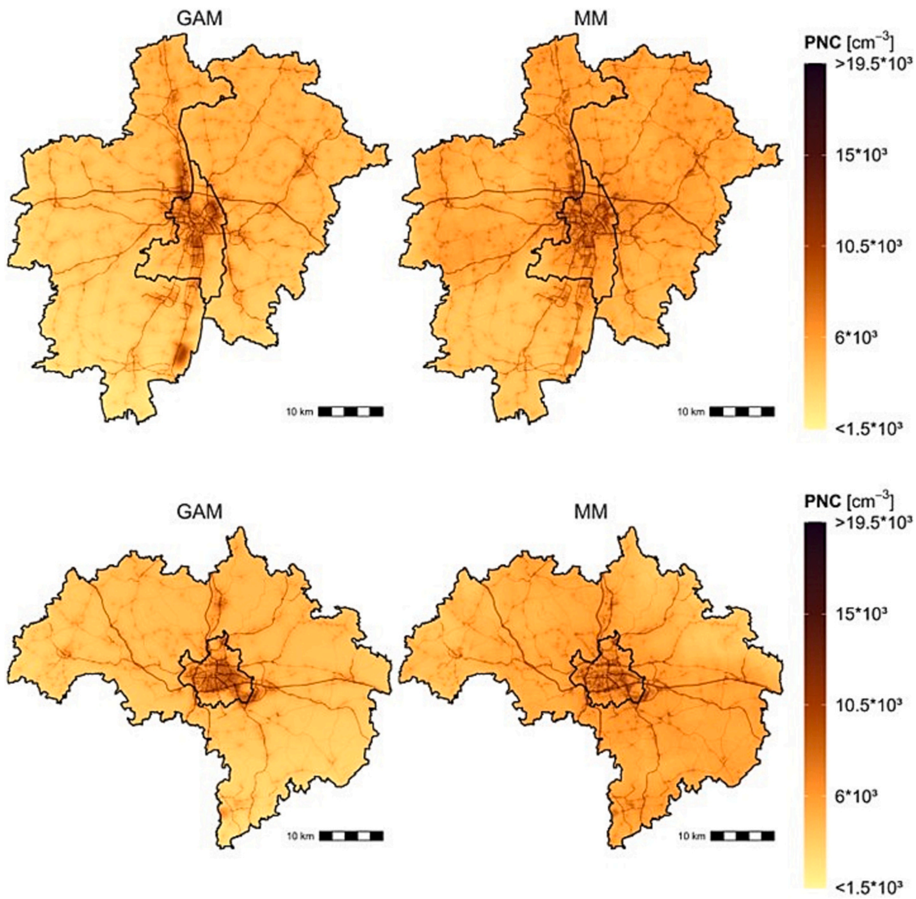

Fig. 1. Annual total particle number concentration (PNC) at a 50-m resolution predicted with a generalized additive model (GAM, left) and a mixed model (MM, right) for the Augsburg (top) and Regensburg (bottom) area in 2017.

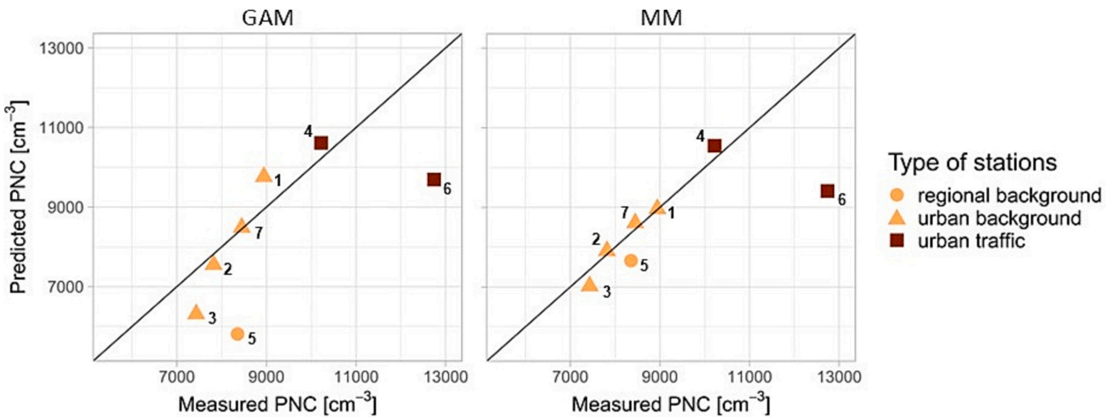

Fig. 2. Scatterplots and linear fits of measured vs predicted long-term annual particle number concentration (PNC) for the generalized additive model (GAM, left) and the mixed model (MM, right) at the monitoring sites in Regensburg (numbers refer to locations in Supplemental Figs. S2 and S5).

Since UFP or PNC routine measurements are still limited and high-quality stationary measurements costly and labour-intensive, most recent approaches used mobile monitoring or short-term stationary measurements or a combination of both (Jones et al., 2020; Kerckhoffs et al., 2021; Kerckhoffs et al., 2019; Patton et al., 2015; Rahman et al., 2020; Simon et al., 2018; van Nunen et al., 2017; Yang et al., 2020; Zalzal et al., 2019). In addition to a fine-scale spatial resolution, they partly also intended to predict short-term temporal variation by incorporating temporal (e.g., meteorology) in addition to spatial predictors. However, due to the increased complexity, the model performance was generally lower with adjusted R^2 ranging from 0.18 to 0.68 for hourly values compared to models that focused on long-term concentrations and spatial variation only (adjusted R^2: 0.28–0.89) (Blanco et al., 2023; Jones et al., 2020; Rahman et al., 2020; Simon et al., 2018; Yang et al., 2020). These are lower or comparable to our adjusted and LOOCV R^2 of 0.90 and 0.76 for the GAM and 0.91 and 0.86 for the MM.

Several studies also investigated the transferability of PNC LUR models to one or several regions (Ma et al., 2024; Patton et al., 2015; van Nunen et al., 2017; Yang et al., 2020; Zalzal et al., 2019). While most of them reported a reduced performance for the transferred regions, a study from Canada showed that transferability worked well between two cities with similar geographical characteristics, with only a minor decrease in model fit and predictive skill (Zalzal et al., 2019). Although we did not develop a separate model for Regensburg, our external validation with new measurements showed good agreement with the transferred predictions. This indicates that the two study regions have comparable characteristics. However, our study also shows that validation is needed to establish transferability and to identify local sources which can be potentially captured by additional predictors.

Strengths of our study include i) high-quality and standardized measurements for model prediction and external validation; ii) comparably long measurement periods at each site covering the warm, cold and intermediate seasons and temporal adjustment by the continuously running reference site to derive long-term annual averages; iii) a large amount of heterogenous and highly resolved spatial predictors; and iv) a standardized and well-established modelling routine that we extended by a temporal component comparing two different modelling approaches. A major limitation is the comparably low number of sites, although we added further measurements compared to our previous model (Wolf et al., 2017). The low number hampered us from applying more advanced modelling (e.g., machine learning approaches) and validation (e.g., hold-out validation) techniques which have been reported to inflate the predictive ability (Dong et al., 2021; Ma et al., 2024; Wang et al., 2013). Further, our predictions tended to underestimate the true concentrations, specifically at traffic sites. Thus, we expect very good agreement at urban and rural areas outside hotspots, but there may be an underestimation of the true exposures at hotspots. This underestimation may bias health effects towards the null in subsequent epidemiological analyses.

5. Conclusions

The two LUR models resulted in similar predictions and captured intra-city spatial patterns and city-rural gradients very well. Our data show that models from one city can effectively be transferred to another city if the geographic features and spatial variability of the predictors across the two study areas are comparable. However, to evaluate the predictive capability in a novel study area, additional validation measurements are recommended.

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CRediT authorship contribution statement

Marco Dallavalle: Writing – original draft, Visualization, Methodology, Formal analysis, Data curation. **Josef Cyrys:** Writing – review & editing, Funding acquisition, Conceptualization. **Susanne Sues:** Writing – review & editing, Resources. **Simonas Kecorius:** Writing – review & editing, Resources. **Susanne Breitner-Busch:** Writing – review & editing, Project administration, Funding acquisition, Conceptualization. **Regina Pickford:** Writing – review & editing, Funding acquisition, Conceptualization. **Alexandra Schneider:** Writing – review & editing, Funding acquisition, Conceptualization. **Annette Peters:** Writing – review & editing, Supervision, Methodology, Funding acquisition, Conceptualization. **Kathrin Wolf:** Writing – original draft, Supervision, Methodology, Funding acquisition, Formal analysis, Data curation, Conceptualization.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

Marco Dallavalle reports financial support was provided by Bavarian State Ministry for the Environment and Consumer Protection. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.uclim.2025.102644.

Data availability

Data can be made available on reasonable request.

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Glossary

BKG: Federal Agency for Cartography and Geodesy
DWD: German Meteorological Service
GAM: Generalized additive model
KORA: Cooperative Health Research in the Region of Augsburg
LfU: Bavarian Environment Agency
LOOCV: Leave-one-out cross-validation.
MM: Linear mixed effects model
NAKO: German National Cohort
NO_2: Nitrogen dioxide
OSM: Open Street Map
$PM_{2.5}$: Particulate matter <2.5 μm
PNC: Particle number concentration
UFP: Ultrafine particles
ULTRA3: Project "ENVIRONMENTAL NANOPARTICLES AND HEALTH: Exposure, Modeling and Epidemiology of Nanoparticles and their Composition within KORA"