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Assessing alternative population size proxies in a wastewater catchment area using mobile device data.

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20 GRAPHICAL ABSTRACT

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23 ABSTRACT

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25 Modeling and prediction of a city's (Oslo, Norway) daily dynamic population using mobile device-based 26 population activity data and three low cost markers is presented for the first time. Such data is useful 27 for wastewater-based epidemiology (WBE), which is an approach used to estimate the population level 28 use of licit and illicit drugs, new psychoactive substances, human exposure to a wide range of pollutants, 29 such as pesticides or phthalates, as well as the release of endogenous substances such as oxidative 30 stress and allergen biomarkers. Comparing WBE results between cities often requires normalization to 31 population size, and inaccuracy in the measured population can introduce high levels of uncertainty. In 32 this study mobile phone data from 8-weeks in 2016 was used to train three linear models based on 33 drinking water production, electricity consumption and online measurements of ammonium in 34 wastewater. The ammonium model showed the best correlation with an R-squared of 0.88 while 35 drinking water production and electricity consumption showed more discrepancies. The three models 36 were then re-evaluated against 5-week of mobile phone data from 2017 showing mean absolute errors 37 <10%. The ammonium-based estimated mean annual population for Oslo in 2017 was 645 000 38 inhabitants, 4% higher than the "de jure" population reported by the wastewater treatment plant. Due 39 to changing conditions and seasonality, drinking water production underestimated the population by 40 27% and electricity consumption overestimated the population by 59%. Therefore, the results of this work showed that the ammonium mass loads can be used as an anthropogenic proxy to monitor and 41 42 correct the fluctuations in population for a specific catchment area. Furthermore, this approach uses a 43 simple, yet reliable indicator for population size that can be used also in other areas of research.

45 INTRODUCTION

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Wastewater-based epidemiology (WBE) is an established tool to estimate the population level exposure to a wide range of pollutants (i.e. pesticides¹, phtalates², phosphorus flame retardants³) by the quantitative measurement of endogenous and exogenous biomarkers excreted by humans in wastewater⁴. Furthermore, WBE already complements existing epidemiology-based estimation techniques of illicit drug use⁵⁻⁶ and more recently has provided human exposure estimates of oxidative stress⁷ and allergen biomarkers⁸. Future strategies have also been discussed to assess the measurement of population health markers and biological markers of microbial exposure and disease⁹.

54 WBE has the potential to provide near real-time information on community exposure to chemicals in 55 form of mass loads that can be used for monitoring purposes, and has also been proposed as a potential 56 early warning system (EWS) tool ¹⁰⁻¹¹. Although WBE provides evidence-based and objective data on 57 human exposure to a range of chemicals, it is still subject to a number of uncertainties. Much of the 58 uncertainty lies in correction factors used to normalize the levels of biomarkers measured. This 59 normalization is essential for allowing comparisons to be made with other geographical locations or 60 during other time intervals ¹².

The assessment of the population size attached to the wastewater treatment plant (WWTP) is crucial and its uncertainty has been estimated to be between 7 and 55% ¹². In a recent study, mobile devicebased population activity (MPA) patterns were used to calculate real-time population within a certain community ¹³. Using this technology, Thomas et al have confirmed that the uncertainty for the assessment of a population within a certain WWTP can be as high as 55%, mainly due to the temporal fluctuations of the population.

67 Two broad population estimates have been defined and used in recent years ¹⁴: de jure (most 68 straightforward and based on the residence census data served by the WWTP) and *de facto* (actual 69 contributors to the system). The first approach does not provide information on whether people are 70 actually within the catchment area throughout the investigation, or are indeed elsewhere. The second 71 approach uses a proxies (such as chemical markers) that reflect the number of individuals actually using 72 the wastewater system during the time of the investigation. However, the proxy may not consider 73 wastewater loss or infiltration and are more vulnerable to additional limitations such as confounding 74 factors (i.e. industrial discharges) ¹⁵.

75 The introduction of a population measure as a *de facto* proxy could significantly diminish uncertainty 76 associated with population normalization of WBE data. To date, the use of MPA patterns has been 77 shown to be the most accurate option for population estimation. However, these resources are not 78 always available and may present a high cost ¹³. Therefore, the use of either endogenous or exogenous 79 substances in the wastewater as population biomarkers (PB) is seen as the better solution in the long 80 term. Ideal PB candidates must (a) be excreted at constant levels and by a representative percentage 81 of the population, (b) be stable and have no affinity to particle matter, (c) be quantifiable in wastewater 82 and (d) be easily analysed and at low cost 16 .

Cholesterol, creatinine, coprostanol and some prescribed pharmaceuticals have been proposed as human specific PB among others ¹⁶⁻²⁰. Lai et al. used a population model based on the measurement of 14 chemicals during a 311-day study to estimate the *de facto* population and compare it against the *de jure*. This study showed on average 32% higher population estimate using the *de facto* model over the

87 *de jure*¹⁵.

Hydrochemical parameters (exogenous substances), such as biological oxygen demand, chemical 88 89 oxygen demand, nitrogen or phosphorus have also been used to estimate the population ²¹⁻²². Been and colleagues used a population-estimate model based on the analysis of ammonium (NH_4^+) derived 90 from a Swiss survey carried out in a 4-year study in 86 WWTPs. The linear model predicted a population 91 equivalent of 8.1 \pm 0.4 g day¹ of ammonium, and using the online measurements of this proxy the 92 authors could quantify weekly and seasonal (i.e. summer holidays) fluctuations in the population ²³. 93 Ammonium (NH4⁺) is present in wastewater as an hydrolysis product of urea ²⁴, and although this is not 94 95 specific to human urine, this marker has been shown to be less influenced by wastewater composition 96 than other parameters. Ammonium is therefore regarded as a good potential PB, especially for 97 predominantly domestic catchments with small industrial discharges²³.

98 Electricity consumption and drinking water production could potentially be considered as another 99 alternative to PB. Both are relatively easy to measure and can theoretically represent the amount of 100 people in a specific area, specially within areas with minor industrial activity. Yet, the major concern 101 regarding these two measurements is the impact of the seasonal variations (i.e. electricity consumption 102 summer vs winter). To the best of our knowledge, none of these data have been previously assessed as 103 a population proxy.

Therefore, the scope of this study was to assess the suitability of three different proxies for the population normalization of WBE data from Oslo (Norway). The three candidates were drinking water production, electricity consumption, and online measurements of ammonium in wastewater. The three proxies were studied against MPA data. Models for a simple, yet reliable indicator for population size, were created with data obtained in an 8-week period in 2016 and tested in a 5-week period in 2017. Finally, the uncertainty was assessed, and population estimates were derived for each of the three models.

111 MATERIALS AND METHODS

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113 Wastewater Treatment Plant and Flow Rates

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The total length of the sewer line connected to VEAS WWTP is about 42.3 km and connects 29 pipelines. 115 116 The residence time in the sewer system, defined as the average time the sewer takes from the 117 households to the treatment plant, has been reported as 5 hours (see www.veas.nu for further details). VEAS treats sewage for approximately 600 000 people (607 651, 615 332 and 619 673 inhabitants 118 estimated in 2015, 2016 and 2017 respectively) of which the city of Oslo contributes about 70.5% and 119 120 the adjoining four different municipalities (Asker, Bærum, Nesodden and Royken) represent the other 121 29.5% (Figure S1). Wastewater flow rate is measured at the end of the plant using a Khafagi Venturi 122 flume with level sensor installed in the outlet channel. Time-adjusted hourly wastewater flow 123 measurements were used to calculate the ammonium mass loads in 2016 and 2017. See S.I. for more 124 information about VEAS WWTP configuration.

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126 Ammonium Measurements and Calculations

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Ammonium concentrations were measured online using a Lange Amtax SC Filtrax (Hach). The one-hour average ammonium measurements (the system logs every 2 seconds) were performed in the

sedimentation tanks, right after the screens and the aerated grit chamber. VEAS WWTP has several 130 131 ammonium devices registering results, and the data is averaged only from those operating adequately 132 (i.e. passing the calibrations). Hourly ammonium data were acquired and reported from January 1st 2016 until December 31st 2017, with the exception of the period from September 9th to 12th in 2017 133 due to system maintenance. VEAS WWTP presents two different lines that recirculate ammonium 134 135 affecting the original concentrations coming downstream into the inlet (see Figure S2). The first line, referred as "wash water", is returning water used to wash the nitrification filters in the nitrification 136 137 tanks and it represents approximately the 5% of the total inlet flow annually while the second, referred 138 as "stripped filtrated water", comes from the ammonia stripping tanks and contains filtrated water 139 extracted from the sludge.

140 The wash water is returned to the main pipe in the inlet while the stripped water is returned in higher 141 concentration but lower flow rates right before the sedimentation tanks. Stripped filtrated water 142 introduces approximately 3% extra of ammonium into the stream every year and the omission of this 143 input would imply and average error of approximately 12%. Furthermore, the continuous changes in 144 the concentration are sometimes noticeable and would affect the final ammonium loads significantly 145 (i.e. minimum and maximum recirculated stripped ammonium in 2017: 150 and 1270 kg day⁻¹). 146 Ammonium concentrations from both washing water and stripped filtrated ammonia were measured 147 in the laboratory with an ammonia ion selective electrode approximately once per week. Average 148 ammonium mass loads obtained from the composite samples collected from the washing water and 149 stripped filtrated tanks were then used to create daily correction factors in order to subtract the 150 ammonia loads originated from the recirculation system as shown below:

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$$k_{\text{amm}} = 1 - \frac{L_{amm} + L_w + L_{st}}{L_{amm}}$$

where k_{amm} is the ammonium correction factor, and L_{amm} , L_w , and L_{st} the 24-hour average ammonium loads measured from raw wastewater entering in the plant, wash water line and stripped filtrated water line respectively. Subsequently, the correspondent daily correction factors were applied to each of the 24 ammonium measurements registered every day. The unknown dynamics behind the mass loads of these measurements, especially L_w and L_{st} due to its low resolution, has not been assessed. However, the use of the k_{amm} will decrease the potential underestimation mentioned above.

Ammonium concentrations are measured at the entrance of the sedimentation tanks while the flow meter is installed in the outlet pipe. The time delay between these two locatons is of about 1 hour. Therefore, ammonium mass loads were calculated by multiplying the wastewater flow rate by the ammonium concentrations measured 1 hour later. Furthermore, 24-hour ammonium mass loads averages were multiplied by k_{amm} and presented in kg h⁻¹ to be compared with the MP data.

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164 Mobile Device-Based Population Data

The data source used to generate the dynamic population weighting in this study was a passive network signaling monitoring system. This monitoring system extracts all signaling data generated by handsets interacting with the mobile phone network provider, Telenor's network. The measurements were aggregated, going from cells to base station, providing a single count per geographical point. In this way the end user was protected from re-identification by inference.

170 The exact catchment boundary for VEAS WWTP was provided by the five different municipalities. The 171 area was then aligned with a subset of 22 000 cells grouped into 1 500 base stations within the 172 catchment boundary distinguishing between those "inside vs outside" VEAS catchment area. Hourly

- 173 measurements (1 snapshot per hour) of the mobile devices within the greater Oslo region were
- 174 collected for two different periods: 8 weeks between May 30th and July 31st 2016 and a 5-week time,
- 175 from June 15th until July 19th in 2017. The hourly MPA data was firstly used to align the ammonium data
- and then to create the daily average estimates from 00:00 to 00:00 to be used for the lineal regression
- 177 models. The MPA trends and general information can be found elsewhere in Thomas et al^{13} .

178 Drinking water production and electricity consumption data

- 179 Daily drinking water production data for Oslo in 2016 and 2017 were provided by Oslo Municipality
- 180 Water and Sewage Administration (VAV) whereas the hourly electricity consumption data for Oslo
- during the same period of time was obtained from Statnett. Both datasets provide information for the whole Oslo region and therefore are not aligned properly with the VEAS catchment area, nor the
- 183 Telenor data.
- Drinking water production do not consider water leakage nor percentages of water used for households, industry or commerce. The total annual production in 2016 was 95.5 mill m³, out of which approximately the 3% was sold to other municipalities, 4% unregistered and public consumption, 6% for garden watering, 17% use for commerce, 40% for households and the remaining 30% was accounted as water loss. Population estimates for the drinking water network provided by VAV were 647 676 and 658 390 inhabitants in 2015 and 2016 respectively. Daily drinking water production averages were obtained in m³ day⁻¹ and directly compared against the MPA data.
- Electricity consumption is heavily dependent on the season and weather conditions. In 2016, the net consumption in Norway was 44% for manufacturing and mining, 34% for private households and agriculture and 22% for construction and other services ²⁵. Yet, this proxy may still reflect the population patterns during periods of time when the season/weather is stable. High resolution electricity consumption data were obtained as hourly measurements (MWh/h) estimated from the aggregation of continuous data, subsequently averaged by day and compared against the daily MPA data.

197 Data Alignment and Linear regression models

- 198 Daily average MPA data was correlated with electricity consumption, drinking water production, and 199 mass loads of ammonium. Both the time series of electricity consumption and drinking water 200 production were temporally aligned with the mobile-phone data. However, a Cross Correlation function (CCF) was used to calculate the delay between the time ammonium is excreted in the urine and the 201 202 measurement at the wastewater treatment plant. In particular, we used the CCF to estimate the correlation between the hourly time series of the mobile device-based population and of the 203 204 ammonium measurements, at different time lags (with 1 hour resolution). The time lag at which the 205 cross-correlation function was maximized was subsecuently applied into the ammonium hourly data to 206 correct the delay.
- A linear model was used to firstly assess the correlation between the daily mobile phone data and theother proxies in the period between June and July 2016.

209 $Y = \alpha + \beta X_i$

- where X_i represents the three proxies: drinking water (X_w) , electricity consumption (X_e) or ammonium
- 211 level (X_a). The three linear models trained on 2016 data have been used to validate the approach and
- 212 predict the population using MPA data from June and July 2017. The models' predictions were
- evaluated against a new mobile phone dataset from 2017 and compared in terms of mean absolute
- 214 percentage error (MAPE):

215 MAPE =
$$\frac{1}{n} \cdot \sum_{i=1}^{N} \left| \frac{Y_i - \hat{Y}_i}{Y_i} \right|$$

where Y_i is the true value (mobile data 2017) and \hat{Y}_i the predicted value for electricity consumption, drinking water production and ammonium mass loads. This function compares the residual to the observed values, providing a standardized measure to evaluate the different models. We used a LOESS (locally weighted scatterplot smoothing) non-parametric function to highlight the trend of the daily population estimates in Oslo for the three models (drinking water, electricity and ammonium) between 01/01/2017 and 31.12.2017.All statistical analyses were performed using the program R, version 3.3.2 (https://www.r-project.org/).

223 RESULTS AND DISCUSSION

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225 Ammonium Data Alignment

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227 The mean lag between the households located within the VEAS catchment area and the entrance of 228 the sedimentation tank where ammonium was measured was best estimated using a CCF between the 229 MPA data and the ammonium measurements in wastewater at different temporal lags (Figure 1). The 230 CCF analysis showed that the correlation between the time series of the mobile data and that of the ammonium concentration are positively correlated, with the highest correlation occurring at lag=-4 231 232 hours. This means that an increase (decrease) of the value of the mobile data activity is associated with 233 an increase (decrease) in the ammonium level 4 hours later. Therefore, all further analysis were 234 performed using 24-hour MPA data against a 4-hour ahead ammonium data (i.e. mobile-derived data 235 from 00:00 to 00:00 and ammonium data from 04:00 to 04:00).



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Figure 1. CCF mode using hourly measures of ammonium (kg h-1) and mobile-derived data to best fitting the mean lag between
 the households and the WWTP.

239 Electricity, Drinking Water and Ammonium in 2016: Training of the Models

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241 MPA data between June and July 2016 were used to train the three linear models with the daily average 242 drinking water production in Oslo, electricity consumption in Oslo and ammonium measured in the

243 VEAS catchment area (see model details in Table 1; Figure 2a,b,c).

Average water production during the monitoring study (June-July 2016) was 261 489 m³ day⁻¹. Drinking 244 245 water production was gradually decreasing during the studied period. The highest drinking water production occurred the first day of the study (Wednesday 01/06/2017, 323 970 m³ day⁻¹) while the 246 lowest production was recorded in the middle of July (Wednesday 16/7/2016, 227 663 m³ day⁻¹). The 247 weekly patterns (presented as ratio weekend/week day, considering weekend as Saturday and Sunday) 248 249 show a reduction of water production during the weekends of about 5%. The MPA data and the water production data show a good linear relationship in the low and high interval, less in the mid-range (see 250 251 linear regression in Figure 2a). With an R-square of 0.6, the main discrepancies are potentially related 252 with the different water usage during the summer season. Furthermore, the linear model estimates 253 work well during the first two weeks but after the third weekend the discrepancy in term of population 254 underestimation is noticeable. For the last two weeks, the estimates seem to invert the tendency and 255 it overestimates the real population by approximately a 20%. Furthermore, the drinking water 256 production model estimate does not seem to follow a clear weekly pattern.

Model	Coefficient	Value	95% CI	p-value	R-squared
	α	-342600	-524895; -160365	0.0004	0.60
Drinking water	β	3.2	2.5; 3.9	3.86e-13	
Electricity	α	-464945	-614298; -315591	5.64e-08	0.75
consumption	β	364	308; 420	< 2e-16	
	α	144079	108864; 179294	2.95e-11	0.88
Ammonium	β	2385	2157; 2612	< 2e-16	

Table 1. Coefficient estimates and R-squared for corresponding linear models for drinking water production (X_w), electricity consumption (X_e) and loads of ammonium (X_a).

259 The average electricity consumption in Oslo during the training study (June-July 2016) was 2 652 260 MWh/h and followed the same pattern described above for drinking water with a decrease in consumption from June to July of about 11%. The highest consumption peak was recorded in June 10th 261 while the minimum consumption occurred the 24^{th} of July (3 498 -1 755 MWh/h). For this case, the 262 263 weekly pattern was clearer and the weekend "drop" was more intense than for MPA data, probably 264 due to the work/industry inactivity during the weekend. The linear model in this case showed a better fitting compared with the drinking water production and the R-squared showed a satisfactory 0.75. The 265 influence of the daily work activity/industry in the electricity consumption seems to affect the model 266 slightly overestimating population during the week days and underestimating during the weekend. 267 268 However, the overall model seems to provide a reliable population prediction and proper trend (Figure 269 2b).



Figure 2. Linear regression models for the estimation of population based on the daily MPA data versus a) drinking water
production b) electricity consumption and c) ammonium measures at the VEAS WWTP in 2016 (above) and comparison of the
MPA data and model predictions (below).

The best linear fit within the three studied population proxies was obtained for the ammonium mass loads (R-squared = 0.88). The average ammonium mass loads during the 2-month study were 149 kg hr⁻¹ with a maximum and minimum mass loads of 228 and 89 kg hr⁻¹ respectively on June 14th and July 17th. Monthly ammonium mass loads averages in 2016 decreased 37% from June to July in agreement with the MPA data which showed 31% decrease. Furthermore, both intra and inter day trends correlate very well and the estimate lines showed in Figure 2c are overlapped most of the time indicating an extremely good prediction.

Electricity, Drinking Water and Ammonium in 2017: Validation of the Models

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The models obtained from the 2016 training dataset were subsequently applied using new data obtained from 2017. Figure 3 shows the performance of the models by comparing the population predictions for 2017 against the actual MPA population during the same period. Furthermore, the models were compared in terms of MAPE providing relatively good values below 0.1 (Table 2).

288 The drinking water production model with a MAPE of 0.0875 provided good average estimates during 289 the validation study in 2017 (drinking water: 450 000 inhabitants vs MPA: 470 000 inhabitants) 290 however, it failed defining the trends over the time. Electricity population model showed the lowest MAPE (0.0792) and provided a proper declining trend. However, in this case it showed a tendency of 291 overestimating the population during the weekdays and underestimating during the weekend. 292 Furthermore, electricity is the proxy with higher probabilities of getting affected but confounding 293 294 factors, especially in winter time. Although the MAPE was not the best (0.0879), the ammonium mass 295 loads model showed consistent estimates throughout the validation study. However, results 296 highlighted in Figure S3c show a potential sistematic deviation of the new data from 2017 compared with the regression line estimated with the data from 2016. This variation could be linked with the 297 298 recirculated ammonium water at VEAS WWTP during that period of time leading to a slightly overestimation of the population. Yet, the error is below 10% and the estimates perfectly describe the 299 trend during the 5-week study. 300

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Additionally, the potential gain of prediction power was assessed by combining the three proxies (see
 Table S1 and Figure S4). The new full model slightly outperformed the ammonium training model with

an R-square of 0.93 and the electricity validation model showing a MAPE of 0.074, being 6% lower. The

effect of drinking water consumption in the full model was found to be not significant. This approach

306 can improve and optimize the selection/rejection of new proxies by comparing its significance in the

307 model versus the effort/cost of obtaining the data.

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Figure 3. Population estimates using the A) drinking water production, B) electricity consumption and C) ammonium mass loads
 data from 2017 (red line) against the 2017 MPA data over the time.

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Model	MAPE
Drinking water production	0.0875
Electricity consumption	0.0792
Ammonium mass loads	0.0879

313 Table 2. Mean absolute percentage error for the different population prediction models for 2017.

314 Total Annual Population Estimates

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Annual population estimates for 2017 were calculated using the three models (Figure 4). It should be noted that the models were trained only on the summer month of June and July 2016. Therefore, when providing annual estimates, the trends were affected by confounding factors such as high-water usage in summer, high electrical consumption in winter or heavy rain events that might affect the wastewater flow rates.

321 Mean drinking water production in 2017 was 245 337 m³ day⁻¹ with a relative standard deviation (RSD) of 6% and minimum and maximum daily mean productions of 204 275 and 275 580 m³ day⁻¹ in April 322 323 and February respectively. The annual population average estimate was 450 000 inhabitants which 324 looks far below(-32%) from the 660 000 estimated in January 2016 for Oslo by Statistics Norway 325 (https://www.ssb.no/en). Water usage during the calibration period seemed to be relatively higher than the rest of the year which resulted in an underestimation of the annual population. Although the 326 327 two datasets do not represent the same two geographical boundaries, the high differences on the 328 estimates seem to indicate that the model do not perform well under changing conditions (seasons). 329 Yet, the annual trend (Figure 4) captures the changes of population during the main festivities such as 330 Easter, summer season or Christmas.

Average electricity consumption in 2017 was 4 166 MWh/h which resulted in an average estimate of 1 332 333 050 000 inhabitants, 59% larger than the official estimates from Statistics Norway. The high variability 334 of the consumption (RSD = 30%, min = 2 162 MWh/h, max = 6 862 MWh/h) was driven by the drastic changes of temperature in Oslo during winter and the consumption of electricity for heating. The 335 average temperature in Oslo in January 2017 was 0°C while in July was 16°C (see Figure S5). Although 336 337 both drinking water production and electricity consumption seemed to be adequate proxies during the model calibration, the annual estimates indicate that those are biased towards the different seasons 338 339 and habits of the population in summer and winter. This suggests that models using electricity as a 340 proxy might need to consider the temperature to adjust the higher consumption of the winter period, 341 especially in those countries characterized by strong seasonal variations. Unfortunately, MPA data was 342 not available during winter season for a better model calibration. Therefore, further research needs to 343 be done in order to implement new models that can take into account these additional variables.

The mean ammonium mass loads for the entire year was 216 kg hour⁻¹ with a RSD of 19% and a 344 minimum and maximum mass loads of 86 and 305 kg hour⁻¹ occurring in October and February 345 respectively. Transformed into inhabitants, the average population estimate at Oslo's catchment area 346 in 2017 was 645 000 inhabitants, 4% higher than the "de jure" population reported by VEAS WWTP 347 348 based on census data and catchment boundaries (620 000 inhabitants estimated in 2017). Furthermore, 349 the model computed a population equivalent of 8,04 grams of ammonium per day and per inhabitant 350 in Oslo. This estimate is in agreement with a previous Swiss study that used ammonium load averages from 86 WWTPs and a nationwide survey to estimate a population equivalent of 8.1 grams ²³. 351 352 Therefore, ammonium mass loads demonstrated to be the most stable proxy (or less influenced by 353 confounding factors) for population estimates and moreover, since the calibration was performed in a 354 single point during the year, the estimates were the best within the ones used for this study. Yet, further 355 studies on the impact of the temperature, incomplete hydrolysis of urea to ammonium, measurement 356 error and maintenance or temporal dynimics would increase the overall knowledge to decrease the 357 uncertainty of this method when implemented in other locations rather than Oslo.

358 The main drops in population observed throughout the year can be explained by Norwegian national 359 festivities. The first major reduction in population takes place in week 9 during the Norwegian winter holiday. Population estimates during Easter drop approximately 25% and the variability in May is very 360 high due to the large number of festivities and national holidays (1st, 17th and 25th of May). The major 361 decrease during the year occurs during the summer break when the number of inhabitants in July 362 decreased 35% compared with the annual mean, in agreement with our previous work ¹³. The 363 population during the second part of the year seems much more stable except for the small drop at the 364 365 beginning of October due to the Norwegian autumn holiday and the general decrease in December during Christmas time. The decrease in the mass loads of ammonium (and therefore population 366 estimates) measured on October 25th are not in agreement with public holidays or events and therefore 367 might indicate an error on the measurement of ammonium in wastewater. The drop of the estimates 368 in September 9 - 12th was due to system maintenance. 369



Figure 4. Daily population estimates in Oslo for the three models (drinking water, electricity and ammonium) between
 01/01/2017 and 31.12.2017. Red curve is a local regression model (loess curve) fitted to the population estimates to highlight
 the temporal trend over the year. Daily estimates are compared with the annual population estimates provided by VEAS WWTP
 (black dots) and Statistics Norway (SSB – dash dots).

375 Implications and Outlooks

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Recent studies have demonstrated that the estimation of the number of inhabitants present within a WWTP catchment is the major source of uncertainty in WBE ¹³. Furthermore, the main failure is not only related to accuracy of the static population estimate but the variability and different trends during the week or the year. This issue can alter the overall conclusion of the WBE results and therefore there is a need for a solution.

In this study, we assessed and compared for the first time the performance of three "low-cost" population proxies for the correction of the weekly, monthly and annual variability within a determined catchment area. Ammonium mass loads measured at VEAS WWTP were shown to be a proper population proxy for the population-normalization in WBE. Furthermore, drinking water production in Oslo and electricity consumption in Oslo are two promising complementary proxies that need to be further evaluated with supplementary MPA data.

388 Drinking water production and electricity consumption provided good estimates during the test study in summer but failed to perform well for the rest of the year due to the changing conditions. However, 389 390 it also needs to be noted that these two proxies were not perfectly aligned with the MPA data since 391 they were measured from the whole Oslo area and not the specific catchment area. Yet, these data are 392 very accessible and can be used in the future alone or integrated in more complex models such as 393 machine learning. Furthermore, the integration of all the covariates in a single model can be used not 394 only to improve the prediction power but to assess the cost/effectiveness of gathering new data for 395 additional proxies (see S.I.). Complementary yearly data on longer periods of time, such as climate data, 396 would contribute to refine the models presented in this study and improve the population estimates.

397 The results obtained in this work showed that the ammonium mass loads can be used as an 398 anthropogenic proxy to monitor and correct the fluctuations in population for a specific catchment 399 area. This work also points out the complexity and importance of gathering reliable ammonium 400 concentrations from the treatment facilities. These are complex systems and the online measurement 401 instruments for ammonium are normally susceptible to the different processes in the plant that can 402 involve drastic changes in the concentrations. Additionally, the average lag time between the area of 403 study and the WWTP needs to be considered. The data alignment assessed in this study showed a mean 404 4-hour delay from the release of the ammonium in the urine and the measurement at the WWTP and 405 therefore this new outcome can be use in future WBE studies for adjusting the timing of the measured 406 results. The population model created with the ammonium mass loads will also allow for retrospective 407 assessment of all WBE data published during the last years in Oslo.

408

410 ASSOCIATED CONTENT

- 411 Supporting Information
- 412

413 Text, Table S1, Figures S1–S4. This material is available free of charge via the Internet at 414 http://pubs.acs.org.

415 AUTHOR INFORMATION

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426

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435 **REFERENCES**

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437 1. Rousis, N. I.; Zuccato, E.; Castiglioni, S., Wastewater-based epidemiology to assess human 438 exposure to pyrethroid pesticides. Environ Int 2017, 99, 213-220. 439 2. Gonzalez-Marino, I.; Rodil, R.; Barrio, I.; Cela, R.; Quintana, J. B., Wastewater-Based 440 Epidemiology as a New Tool for Estimating Population Exposure to Phthalate Plasticizers. Environ Sci 441 Technol 2017, 51 (7), 3902-3910. Been, F.; Bastiaensen, M.; Lai, F. Y.; van Nuijs, A. L. N.; Covaci, A., Liquid Chromatography-442 3. 443 Tandem Mass Spectrometry Analysis of Biomarkers of Exposure to Phosphorus Flame Retardants in 444 Wastewater to Monitor Community-Wide Exposure. Anal Chem 2017, 89 (18), 10045-10053. 445 4. Gracia-Lor, E.; Rousis, N. I.; Hernández, F. I.; Zuccato, E.; Castiglioni, S., Wastewater-Based 446 Epidemiology as a Novel Biomonitoring Tool to Evaluate Human Exposure To Pollutants. ACS 447 Publications: 2018. 448 5. Thomas, K. V.; Bijlsma, L.; Castiglioni, S.; Covaci, A.; Emke, E.; Grabic, R.; Hernandez, F.; 449 Karolak, S.; Kasprzyk-Hordern, B.; Lindberg, R. H.; Lopez de Alda, M.; Meierjohann, A.; Ort, C.; Pico, Y.; 450 Quintana, J. B.; Reid, M.; Rieckermann, J.; Terzic, S.; van Nuijs, A. L.; de Voogt, P., Comparing illicit 451 drug use in 19 European cities through sewage analysis. Sci Total Environ 2012, 432, 432-9. 452 6. Ort, C.; Bijlsma, L.; Castiglioni, S.; Covaci, A.; de Voogt, P.; Emke, E.; Hernández, F.; Reid, M.; 453 van Nuijs, A. L.; Thomas, K. V., Wastewater Analysis for Community-Wide Drugs Use Assessment. In 454 Handbook of Experimental Pharmacology. Springer, Berlin 2018. 455 7. Ryu, Y.; Gracia-Lor, E.; Bade, R.; Baz-Lomba, J.; Bramness, J. G.; Castiglioni, S.; Castrignanò, E.; 456 Causanilles, A.; Covaci, A.; de Voogt, P., Increased levels of the oxidative stress biomarker 8-iso-457 prostaglandin F2 α in wastewater associated with tobacco use. *Scientific Reports* **2016**, *6*. 458 Choi, P. M.; O'Brien, J. W.; Li, J.; Jiang, G.; Thomas, K. V.; Mueller, J. F., Population histamine 8. 459 burden assessed using wastewater-based epidemiology: The association of 1, 4-methylimidazole 460 acetic acid and fexofenadine. Environment international 2018, 120, 172-180. 461 Choi, P. M.; Tscharke, B. J.; Donner, E.; O'Brien, J. W.; Grant, S. C.; Kaserzon, S. L.; Mackie, R.; 9. O'Malley, E.; Crosbie, N. D.; Thomas, K. V.; Mueller, J. F., Wastewater-based epidemiology biomarkers: 462 463 Past, present and future. *Trac-Trend Anal Chem* **2018**, *105*, 453-469. 464 10. Baz-Lomba, J. A.; Love, A. S.; Reid, M. J.; Olafsdottir, K.; Thomas, K. V., A high-throughput 465 solid-phase microextraction and post-loop mixing large volume injection method for water samples. J 466 Chromatogr A 2018, 1531, 32-38. 467 Alygizakis, N. A.; Samanipour, S.; Hollender, J.; Ibáñez, M.; Kaserzon, S.; Kokkali, V.; van 11. Leerdam, J. A.; Mueller, J. F.; Pijnappels, M.; Reid, M. J., Exploring the potential of a global emerging 468 469 contaminant early warning network through the use of retrospective suspect screening with high-470 resolution mass spectrometry. Environmental science & technology 2018, 52 (9), 5135-5144. 471 12. Castiglioni, S.; Bijlsma, L.; Covaci, A.; Emke, E.; Hernandez, F.; Reid, M.; Ort, C.; Thomas, K. V.; 472 van Nuijs, A. L.; de Voogt, P.; Zuccato, E., Evaluation of uncertainties associated with the 473 determination of community drug use through the measurement of sewage drug biomarkers. Environ 474 Sci Technol 2013, 47 (3), 1452-60. 475 Thomas, K. V.; Amador, A.; Baz-Lomba, J. A.; Reid, M., Use of Mobile Device Data To Better 13. 476 Estimate Dynamic Population Size for Wastewater-Based Epidemiology. Environ Sci Technol 2017, 51 477 (19), 11363-11370. 478 14. Daughton, C. G., Monitoring wastewater for assessing community health: Sewage Chemical-479 Information Mining (SCIM). Sci Total Environ 2018, 619-620, 748-764. 480 Lai, F. Y.; Anuj, S.; Bruno, R.; Carter, S.; Gartner, C.; Hall, W.; Kirkbride, K. P.; Mueller, J. F.; 15. 481 O'Brien, J. W.; Prichard, J.; Thai, P. K.; Ort, C., Systematic and day-to-day effects of chemical-derived

- 482 population estimates on wastewater-based drug epidemiology. *Environ Sci Technol* 2015, *49* (2), 999483 1008.
- 16. Chen, C.; Kostakis, C.; Gerber, J. P.; Tscharke, B. J.; Irvine, R. J.; White, J. M., Towards finding a population biomarker for wastewater epidemiology studies. *Sci Total Environ* **2014**, *487*, 621-8.
- 486 17. Chiaia, A. C.; Banta-Green, C.; Field, J., Eliminating Solid Phase Extraction with Large-Volume
- Injection LC/MS/MS: Analysis of Illicit and Legal Drugs and Human Urine Indicators in US Wastewaters.
 Environmental Science & Technology 2008, *42* (23), 8841-8848.
- 489 18. Brewer, A. J.; Ort, C.; Banta-Green, C. J.; Berset, J. D.; Field, J. A., Normalized diurnal and
- between-day trends in illicit and legal drug loads that account for changes in population. *Environ Sci Technol* 2012, 46 (15), 8305-14.
- 492 19. Daughton, C. G., Real-time estimation of small-area populations with human biomarkers in
 493 sewage. *Sci Total Environ* **2012**, *414*, 6-21.
- 494 20. O'Brien, J. W.; Thai, P. K.; Eaglesham, G.; Ort, C.; Scheidegger, A.; Carter, S.; Lai, F. Y.; Mueller,
 495 J. F., A model to estimate the population contributing to the wastewater using samples collected on
 496 census day. *Environ Sci Technol* 2014, *48* (1), 517-25.
- 497 21. Rico, M.; Andres-Costa, M. J.; Pico, Y., Estimating population size in wastewater-based
- 498 epidemiology. Valencia metropolitan area as a case study. *J Hazard Mater* **2017**, *323* (Pt A), 156-165.
- 499 22. Zheng, Q. D.; Lin, J. G.; Pei, W.; Guo, M. X.; Wang, Z.; Wang, D. G., Estimating nicotine
- 500 consumption in eight cities using sewage epidemiology based on ammonia nitrogen equivalent 501 population. *Sci Total Environ* **2017**, *590-591* (Supplement C), 226-232.
- 502 23. Been, F.; Rossi, L.; Ort, C.; Rudaz, S.; Delemont, O.; Esseiva, P., Population normalization with
- ammonium in wastewater-based epidemiology: application to illicit drug monitoring. *Environ Sci Technol* 2014, 48 (14), 8162-9.
- Rauch, W.; Brockmann, D.; Peters, I.; Larsen, T. A.; Gujer, W., Combining urine separation with
 waste design: an analysis using a stochastic model for urine production. *Water Research* 2003, *37* (3),
 681-689.
- 508 25. Norway, S., Net consumption of electricity, by type and consumer group in Norway. 2018; pp 509 <u>https://www.ssb.no/en/energi-og-industri/statistikker/elektrisitet/aar</u>.
- 510