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Estimation of the NO_2 population exposure in the Northern Harbour district of Malta

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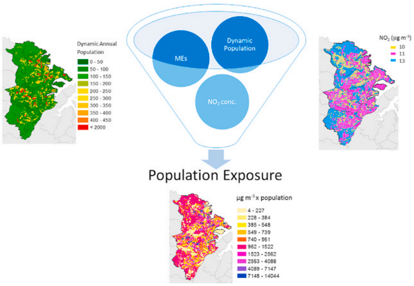
HIGHLIGHTS

- The first study to estimate NO_2 exposure considering population mobility in Malta.
- The highest NO_2 exposure is noted in the home microenvironment.
- The lowest NO_2 exposure is estimated for the traffic microenvironment.
- The NO_2 exposure is 25% higher when using a constant NO_2 concentration for MEs.
- The total NO_2 exposure is 3% less if a static population is used for each time step.

GRAPHICAL ABSTRACT



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ABSTRACT

This study presents an estimate of the total NO_2 exposure in a polluted and densely populated region in Malta, the Northern Harbour district. To estimate the population mobility, we follow a dynamic approach whereby four microenvironments are mapped onto defined Copernicus Urban Atlas 2012 land use classifications. These include the home (ME_home), work (ME_work), traffic (ME_traffic), and other outdoor activities (ME_other) microenvironments. In addition, generic time-activity profiles are used to estimate the hourly mobility in the different microenvironments depending on weekday or weekend profiles. Measured hourly NO_2 ambient concentrations from the air quality network run by the Environment and Resources Authority (ERA) are used in conjunction with the estimated time-activity profiles to calculate the total exposure.

The highest NO_2 population exposure is estimated for ME_home (71%) as people spend the majority of the time in this microenvironment followed by ME_other (15%), ME_work (10%) and ME_traffic (4%), respectively. In addition, we test the sensitivity of the total NO_2 exposure to changes in NO_2 concentrations for different microenvironments. The total NO_2 exposure using infiltration rates to estimate infiltrated outdoor NO_2 concentrations in indoor microenvironments, is up to 25% lower compared to the NO_2 exposure estimated using outdoor NO_2 concentrations for all microenvironments. Results also suggest a decrease of 3% in the estimated NO_2 exposure if a static population is assumed for each microenvironment as opposed to a dynamic one. Exposure assessments such as that presented in this study are essential to aid the development of targeted policies to limit such exposures.

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1. Introduction

Identifying the exposure to indoor and outdoor air pollution is essential as a first step to aid the formulation and implementation of targeted policies to limit air pollution exposure. The World Health Organisation (WHO) as well as the European Environmental Agency (EEA) outline three major outdoor air pollutants most detrimental to health, one of which is nitrogen dioxide (NO_2) (EEA, 2019; WHO, 2005). Nitrogen oxides (NO_X) emissions can be both natural (~23%) and anthropogenic (~77%; Seinfeld and Pandis, 2016). Some examples of natural emissions of NO_X include volcanic emissions and lightning. In contrast, anthropogenic sources of NO_X emissions are mostly related to the burning of fossil fuels for example for heating and power generations as well as vehicle emissions (EEA, 2019; Seinfeld and Pandis, 2016).

NO_2 has several properties which make it an important pollutant to study. Apart from the associated adverse health effects, the absorbing qualities of NO_2 can lead to impaired visibility as well as global climate change at high levels (WHO, 2000). In addition, NO_2 plays a critical role in the photochemical formation of ozone (Monks et al., 2015; Seinfeld and Pandis, 2016); another key outdoor air pollutant resulting in various adverse health effects (e.g. Malley et al., 2017; Turner et al., 2015; WHO, 2013a, 2015). Globally and regionally, efforts have been made to reduce NO_2 concentrations by setting guidelines as well as target and limit values for countries to adhere to (EEA (European Environmental Agency), 2019; US Environmental Protection Agency (EPA), 2017; WHO, 2005).

Several adverse health effects are associated with both long- and short-term exposure to outdoor NO_2. Short-term outdoor NO_2 exposure is related to increased hospital admissions for respiratory diseases (WHO, 2013a), exacerbations of asthma in both children and adults (Guarnieri and Balmes, 2014) as well as all-cause mortality (WHO, 2013). In addition, long-term exposure to outdoor NO_2 concentrations has been associated with new-onset asthma (Guarnieri and Balmes, 2014), lung function issues (He et al., 2019; Royal College of Physicians (RCP), 2016) as well as cause specific (Crouse et al., 2015; Krewski et al., 2009) and all-cause (natural) mortality (COMEAP, 2018). However, evidence related to indoor NO_2 exposure is limited and mostly related to home exposures (Salonen et al., 2019). Studies focusing on offices and schools have suggested high indoor NO_2 concentrations in urban and traffic hotspots compared to rural locations (e.g. Al-Hemoud et al., 2017; Villanueva et al., 2018).

Personal exposure is mostly determined by the air pollutant concentration in different environments where people spend their time as well as the amount of time they spend within each environment (WHO, 2005). Therefore, the exposure cannot simply be assessed by the pollutant levels. This also depends on the number of people exposed to that air pollutant concentration and for how long. Total exposure represents the exposure in all microenvironments both indoors and outdoors (WHO, 2005). Over the past years, several different exposure metrics have been implemented which vary depending on the air pollutant of interest, the study design as well as the health outcome (Dionisio et al., 2016; Özkaynak et al., 2013). Exposure assessments have developed over time and increased in complexity starting with the simplest approach of using measured outdoor air pollutant concentrations from fixed monitoring sites to the most complex approach, that of exposure modelling. The latter more complex approach may include the use of monitoring data, emission data, meteorological data land-use and or topography, time-activity profiles as well as microenvironmental characteristics (Özkaynak et al., 2013; WHO, 2005). Kazakos et al. (2020) have quantified the health burden misclassification because of different exposure approaches in London. The authors estimate a misclassification of 1174–1541 mean predicted mortalities in Greater London Area by using outdoor concentrations as a representative concentration of total exposure (i.e. both indoor and outdoor irrespective of the microenvironment). Furthermore, their study highlights the importance of incorporating different microenvironments as well as increasing the complexity of such models to limit this misclassification of health burden assessments. For this reason, multiple efforts have been made within the science community to shift from the more simplistic exposure estimates to more complex and dynamic exposure modelling techniques (e.g. Gariazzo et al., 2020; Li and Friedrich, 2019; McGrath et al., 2017; Reis et al., 2018; Terry et al., 2014). For example, Li and Friedrich (2019) developed a probabilistic exposure methodology and suggest that the lifelong exposure to $PM_{2.5}$ and NO_2 is also affected by age, gender and socio-economic status. Personal activity diaries and surveys are subject specific and might not capture the true population-level exposure. In addition, such data and surveys are not easily available through national or regional records. To address this issue, Ramacher et al. (2019) developed a novel generic approach to model dynamic population activity in various microenvironments with the use of publicly available data.

In Europe, about 10% of all reporting stations recorded NO_2 concentrations above the annual EU and WHO limit of 40 μg m^{-3} in 2017, primarily at traffic stations (EEA, 2019). Even though NO_2 concentrations and exposures have decreased along the years, around 7% of the urban population within the European Union are exposed to elevated concentrations of NO_2 which exceed the annual limit value compared to a maximum of 31% in 2003 (EEA, 2019; Guerreiro et al., 2014). In Malta, annual mean NO_2 concentrations range from 3.2 μg m^{-3} in the rural background site of Għarb, Gozo to 38.6 μg m^{-3} in the traffic site of Msida. Being part of the European Union (EU) since 2004, Malta is bound by EU regulations on NO_2 limits and target values. The annual limit of 40 μg m^{-3} for NO_2 concentrations was not exceeded in any of the stations in the Maltese Islands between 2009 and 2017. Annual NO_2 exceedances were only recorded in 2008 reaching 60.9 μg m^{-3} in Msida. Even though no exceedances are recorded for NO_2 between 2009 and 2017, no statistically significant reductions are found in Msida across these years (Fenech and Aquilina, 2020).

In this study we focus on the Northern Harbour district of Malta which has the highest recorded levels of NO_2 compared to the other districts and also registers the highest population totals (Environment Resource Authority (ERA), 2018; National Statistics Office Malta (NSO), 2017). The health burden associated with long-term exposure to NO_2 concentrations between 2010 and 2015 has already been estimated (Fenech and Aquilina, 2020). However, the method used by Fenech and Aquilina (2020) considers a static population exposed exclusively to outdoor NO_2 concentrations in conjunction with pre-established concentrations response coefficients in literature (COMEAP, 2015; WHO, 2013). Though extremely limited, exposure assessments in Malta, typically lack spatial or temporal resolutions. In this study we focus on including a dynamic modelling approach to estimate the total population exposure to hourly NO_2 concentrations in 2017 in the Northern Harbour district. Using this novel approach, we estimate the hourly movement of people and in turn study the overall population exposure in the different microenvironments within this district. The Northern Harbour district is characterised by a continuously changing urban fabric with residential and working population totals on the increase for the past five years however, no local policies have been targeted to improve personal exposure within this district as evidence on local personal exposure is lacking. Using a dynamic approach to estimate personal exposure avoids the recruitment of large cohorts of people which would be both time consuming and expensive. To our knowledge this is the first study conducted in Malta to present an estimate of the total population exposure using a generic approach to simulate population mobility across different microenvironments.

The rest of the paper will be divided as follows: first the methods used in this study will be described in Section 2 followed by results and discussion in Section 3 and ending with a summary and conclusion in Section 4.

2. Methods

2.1. Measured outdoor NO_2 concentrations

This study is conducted in the Maltese archipelago with an area of approximately 316 km^2 and a population density if 1380 persons km^{-2} with 93.2% of the population living in the urbanised areas (Worldometer, 2020). The Maltese Islands are divided into a total of six districts: Northern, Northern Harbour, South Eastern, Southern Harbour, and Western districts in Malta and one district representing the islands of Gozo and Comino (Fig. 1). Air pollution concentrations in the Maltese Islands are routinely measured by the Environment and Resources Authority (ERA) which manages the Air Quality Monitoring Station network at four sites in Malta representing the Western (Attard), Northern Harbour (Msida), Southern Harbour (Kordin) and South Eastern (Żejtun) districts and one in Gozo (Għarb) (Fig. 1). Further details on this network are given in Fenech and Aquilina (2020).

The district with the highest measured outdoor NO_2 concentrations and population totals is the Northern Harbour district reaching an annual mean NO_2 concentration of 38.59 μg m^{-3} at the traffic site of Msida between 2008 and 2017 (Fenech and Aquilina, 2020) and a population of 151,664 in 2017 (NSO, 2019). The Northern Harbour district is one of the smallest districts in Malta covering approximately 10% of the land. In addition, no statistically significant trends in NO_2 concentrations between 2008 and 2017 are found at the Msida site (Fenech and Aquilina, 2020) therefore, making this district an important one to study especially in the context of NO_2 exposure. The only measurement site located within the Northern Harbour district is the Msida traffic site. This site typically exhibits high NO_2 concentrations which may overestimate the overall urban background concentrations within the whole district. To avoid the overestimation of NO_2 concentrations at urban background sites, we apply an hourly ratio derived from the relationship between NO_2 concentrations at the traffic site of Msida and those measured at the urban background site of Żejtun. These two sites exhibit a similar diurnal profile albeit lower NO_2 concentrations at Żejtun. At both stations, peak morning and evening NO_2 levels are measured at 7am and 7pm, respectively. Thus, for each hour we scale the Msida NO_2 concentrations by an hourly ratio ranging from 0.23 (at 2pm) to a maximum of 0.54 (at 7am). The NO_2 concentrations used throughout this study are therefore outdoor hourly concentrations measured at the Msida traffic site in 2017 but scaled by an hourly ratio representative of urban background concentrations.

Fig. 1. Definition of the district boundaries across the Maltese Islands. The air quality monitoring stations maintained by the Environment and Resource Authority (ERA) at: Attard (ATD), Msida (MSD), Kordin (KDN), Żejtun (ŻTN) and Għarb (GRB) are also shown.

2.2. Dynamic population exposure

In this section we describe the procedure followed to estimate the population mobility which is based on the novel approach described by Ramacher et al., (2019) as well as the overall total NO_2 exposure in the Northern Harbour district.

2.2.1. Definition of the microenvironments in the Northern Harbour district

To estimate the population mobility within the district we firstly define different microenvironments (MEs). The spatial distribution of these MEs is based on the Copernicus European Urban Atlas Land Use and Land Cover dataset for the 2012 (UA2012) reference year which is the latest validated dataset (Copernicus Land Monitoring Service, 2012b). This dataset is mainly based on Very High Resolution satellite imagery to provide detail on the density of the urban fabric. In addition, the UA2012 dataset incorporates other information such as road networks and other services and utilities. However, although using this dataset does not allow the distinction between for example commercial buildings and homes one can easily separate between for example green areas, urban areas or roads (more details on this dataset can be found in Copernicus Land Monitoring Service (2012a)). In this study we use the same assumptions used by Ramacher et al. (2019) to map the different MEs onto the UA2012 classifications. Using a dynamic approach, the population activity in the Northern Harbour district is assigned to four different MEs: the work microenvironment (ME_work), the home microenvironment (ME_home), the traffic microenvironment (ME_traffic) and the other microenvironment (ME_other). Table 1 lists the different classifications within the UA2012 dataset and the corresponding MEs associated with each classification. The 'discontinuous urban fabric' classification as well as the 'isolated structures' were mapped onto ME_home while industrial, commercial, mineral extractions and construction site classification where assigned to ME_work. As no information is available to distinguish different facilities within a 'continuous urban fabric' classification, based on local general knowledge, the assumption applied in Ramacher et al. (2019) is used and therefore 70% and 30% of this UA2012 classification is assigned to ME_work and ME_home, respectively. Thus, accounting for work environments in more densely built areas. The proportion of employed people in the Norther Harbour district assigned to ME_work was also scaled to match census data (i.e. people living and working in the

Table 1
The UA2012 Land Use Land Cover classification together with the assigned MEs.

Microenvironment	Code	UA2012 Classification
ME_home	11210	Discontinuous Dense Urban Fabric
	11220	Discontinuous Medium Density Urban Fabric
	11230	Discontinuous Low Density Urban Fabric
	11240	Discontinuous Very Low Density Urban Fabric
	11300	Isolated Structures
ME_work	12100	Industrial, commercial, public, military, and private units
	13100	Mineral extraction and dump sites
	13300	Construction sites
70% ME_work/30% ME_home	11100	Continuous Urban Fabric
ME_other	13400	Land without current use
	14100	Green urban areas
	14200	Sports and leisure facilities
	21000	Arable land (annual crops)
	23000	Pastures
	32000	Herbaceous vegetation associations (e.g. natural grassland)
	33000	Open spaces with little or no vegetations (e.g. beaches)
ME_traffic	12220	Other roads and associated land

Northern Harbour district (NSO, 2014)). While these totals represent the majority of the workforce in the Northern Harbour district, employees working within this district but residing elsewhere in Malta are not accounted for. The 'roads and associated lands' classification was assigned to ME_traffic while all other classifications such as 'green urban areas', 'sports and leisure facilities' and 'open spaces' where mapped onto ME_other. The ME_other classification therefore represents outdoor activities excluding those which are traffic related.

The resultant spatial distribution of the different MEs in the Northern Harbour is illustrated in Fig. 2. It can be clearly seen that, following the definitions of the different MEs, this district predominantly consists of two MEs: ME_home and ME_work (Fig. 2a). Overall, ME_home constitutes much of the Northern Harbour district area at 34% followed by ME_work at 32%, ME_other at 24% and ME_traffic at 11% (Fig. 2b).

2.2.2. *Estimation of the population mobility*

Following the mapping of the different MEs onto the Urban Atlas classification, we analyse the population totals within each ME. The population estimates used in this study are those included in the UA2012 dataset as an attribute of the polygon features (Copernicus Land Monitoring Service, 2012b). This way we ensure coherent sources of data and avoid any additional biases. The National Statistics Office (NSO) 2012 population totals for the overall Maltese population (excluding Gozo and Comino) and the Northern Harbour district are 391,087 and 122,954, respectively (Table 2; NSO, 2019). The corresponding population total for the Northern Harbour district for 2017 is of 151,664. To represent the spatial distribution of the 2017 population, we scaled the UA2012 population data by 1.23%. In addition, we made sure that the ratio between the respective towns making up the Northern Harbour district remains unaltered and represents the true population totals of the individual towns, with the highest population in the town of Birkirkara and the lowest in Ta' Xbiex.

To add a temporal dimension to the population within each ME, we assign generic diurnal profiles for the whole population. As no such data is available locally, we use the static population data mentioned above and apply diurnal fractions obtained from Ramacher et al. (2019). The diurnal activity profile is characteristic of a European one (Borrego et al., 2009) with most of the time spent indoors especially during weekdays (Fig. 3a). Peak working European hours (9am – 6pm) are represented by a higher proportion of the population in ME_work reaching a maximum at noon during weekdays (Fig. 3a). This profile is typical of the Maltese Islands with morning rush hours occurring at around 7am followed by a flatter peak at around 7pm. The weekend time-activity profile allows for a greater proportion of the population to venture outdoors with up to 50% in ME_other between 3pm and 5pm an even larger proportion of the population in ME_home in the morning and evening (Fig. 3b). Using these generic time-activity profiles in conjunction with the static population data we can simulate and estimate the hourly population mobility within the different MEs.

Table 2
Comparison between the 2012 and 2017 population estimates from the national Maltese database (NSO, 2019) and the UA2012 dataset (Copernicus Land Monitoring Service, 2012b).

Population	Malta	Northern Harbour District
UA2012	384,610	122,883
NSO (2012)	391,087	122,954
NSO (2017)	442,978	151,664

The total population exposure is then estimated for each hour of every day. This is done by multiplying the spatially and temporally varying dynamic population (described in Sections 2.2.1 and 2.2.2) with the associated measured hourly NO_2 concentrations at the Msida station in 2017. In this study we present the annual average of the hourly NO_2 exposure estimates in the Northern Harbour district.

2.2.3. *Sensitivity of the NO_2 exposure to a varying pollutant concentration in different MEs and a static population*

Throughout the study we assume that all microenvironments are exposed to the outdoor scaled NO_2 concentrations measured at the traffic site of Msida (representative of urban background concentrations). However, NO_2 concentrations in indoor environments can vary

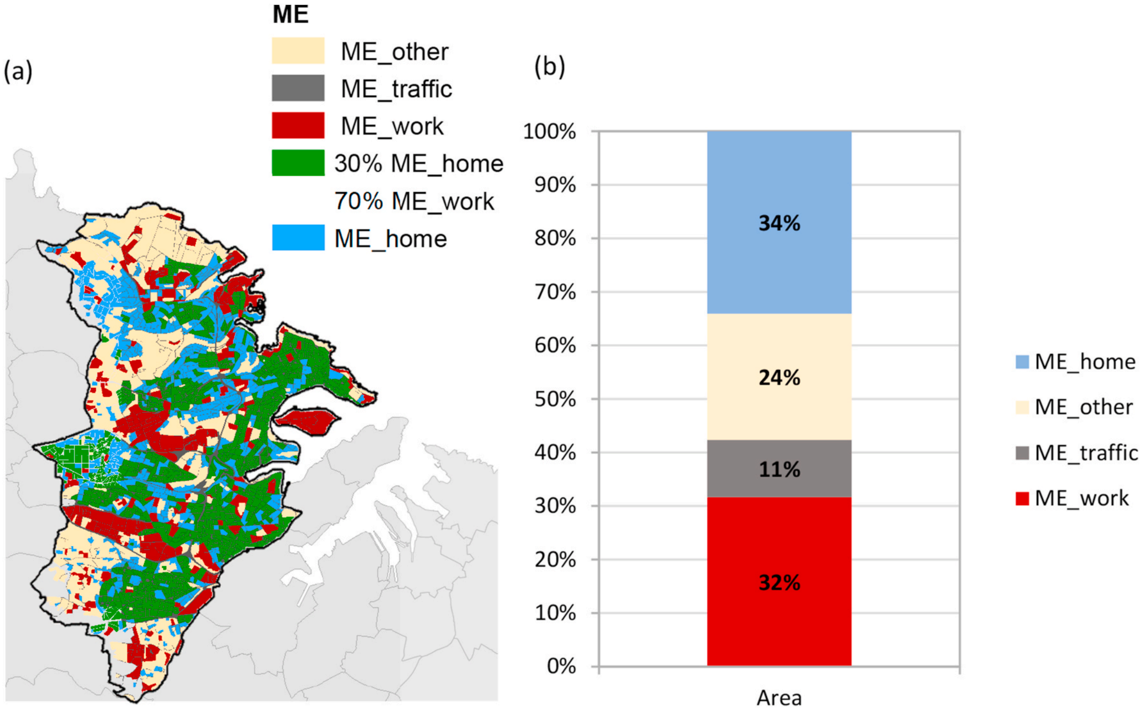


Fig. 2. (a) Spatial distribution of the different microenvironments which are mapped onto the UA2012 classification described in Table 1 and (b) the fraction of land associated with each ME as a function of the total Northern Harbour district area (expressed as a percentage).

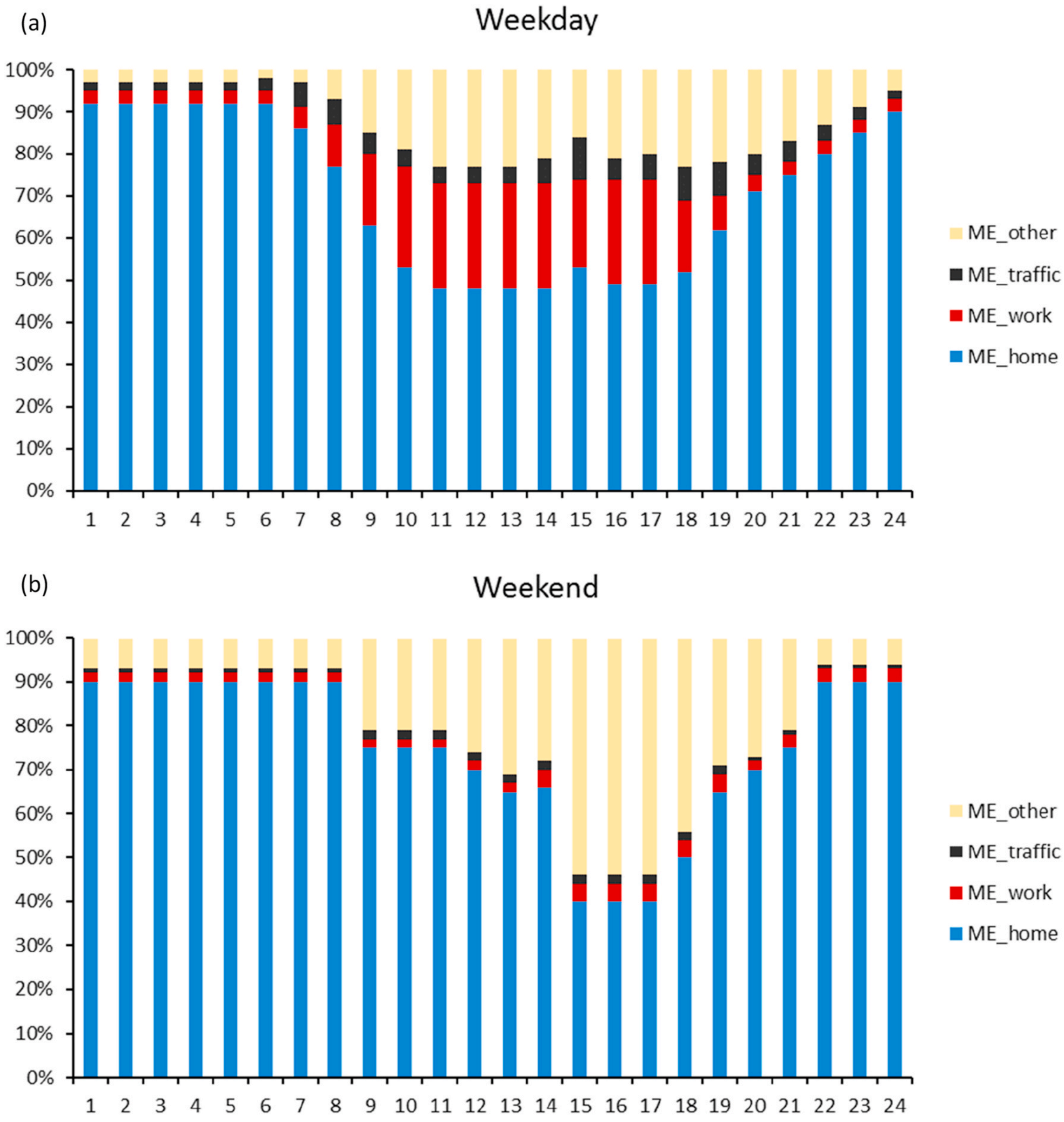


Fig. 3. Generic time-activity profiles to estimate the population mobility during (a) weekdays and (b) weekends.

from the outdoor levels. Some outdoor concentrations may infiltrate indoors and the degree to which this happens depends on several factors such as the building type and materials, ventilation systems as well as outdoor meteorology (e.g. Gaffin et al., 2018; WHO, 2010). In addition, indoor NO_2 concentrations may be higher than those outdoors due to their generation from indoor sources such as cooking and wood burning (Li and Friedrich, 2019). However, wood burning in Malta is not very common and only limited to the colder months mostly using HVAC systems (December to February). To estimate the indoor NO_2 concentrations in ME_home and ME_work which both represent indoor environments, we apply infiltration factors based on values implemented by Ramacher et al. (2019). Unfortunately, local infiltration data is very limited as most local studies focus on the infiltration of other air pollutants such as particulate matter at different size fractions (Vella, 2019) or Volatile Organic Compounds (Hicklin et al., 2018). Limited local evidence suggests low mean air exchange rates in buildings (<3 h^{-1}) with ratios between indoor and outdoor concentrations ranging mostly between 0.5 and 0.8 (Hicklin et al., 2018; Vella, 2019). The infiltration factors used in this sensitivity test for indoor MEs are representative of the Southern European region and range between 0.7 in summer and 0.85 in winter for ME_home and ME_work (Table 3). The rest of the MEs represent outdoor activity profiles and therefore no changes in the ambient concentrations were made in this case (i.e. I/O is set to 1). The indoor/outdoor (I/O) ratios used in this study are broadly consistent with other studies. For example I/O ratios in four elementary schools in Lisbon range between 0.36 and 0.95 with indoor NO_2 concentrations from 15 to 37 μg m^{-3} in spring (Pegas et al., 2011).

We also estimate the total exposure assuming a static population for every time step. To represent a static population the original population

Table 3
Different infiltration factors used to estimate the indoor NO_2 concentrations in each microenvironment during winter and summer (Ramacher et al., 2019).

Microenvironment	Infiltration Factors	
	Winter (Sep–Feb)	Summer (Mar–Aug)
ME_home	0.70	0.80
ME_work	0.75	0.85
ME_other	1.00	1.00
ME_traffic	1.00	1.00

distribution from the UA2012 database was left unchanged for every time step and assigned to ME_home as population totals are based on residential addresses as defined by (Ramacher and Karl, 2020). In addition, when using the static population, we also estimate the total exposure with and without the infiltration factors for the indoor MEs.

3. Results and discussion

The results section is divided into two main parts, firstly we present results of the estimated NO_2 exposure by simulating the movement of people in the different MEs but using the measured outdoor NO_2 concentrations for all MEs (Section 3.1). Secondly, we carry out a set of

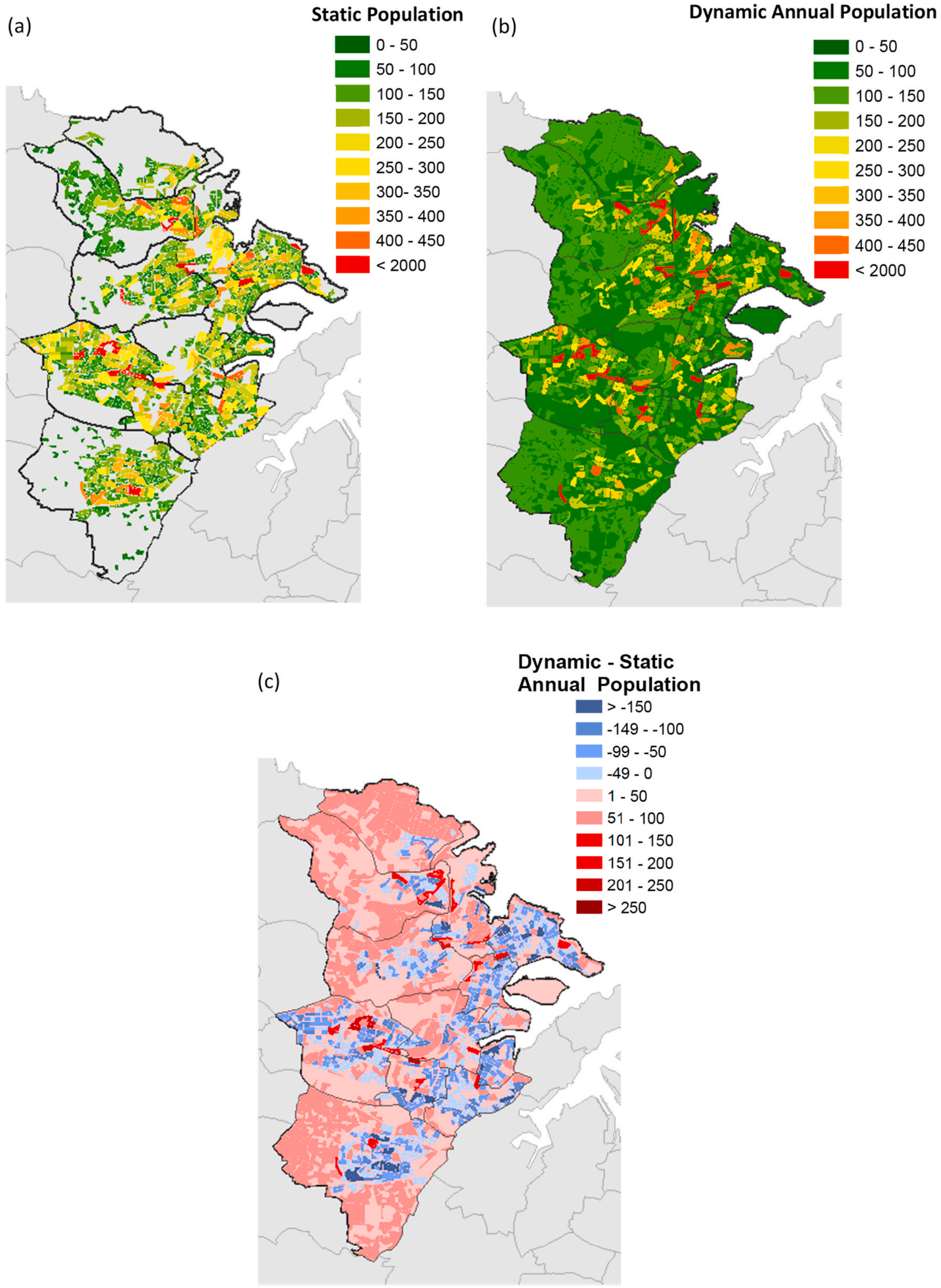


Fig. 4. (a) UA2012 (static) population distribution scaled to 2017 population totals, (b) annual mean dynamic population and (c) difference between the static (UA2012) and annual mean dynamic population (calculated as annual dynamic population – static population).

sensitivity tests to assess the impact of infiltrated outdoor pollutant concentrations into indoor MEs as well as the impact of using a static population approach as opposed to a dynamic approach on the total exposure estimates (Section 3.2).

3.1. *Dynamic population exposure using ambient NO_2 concentrations*

Fig. 4 illustrates the spatial distribution of the original population in the UA2012 dataset scaled to 2017 totals (as described in Section 2.2.2; Fig. 4a) as well as the annual mean dynamic population (Fig. 4b) as estimated using the generic time-activity profiles and MEs definitions described in Section 2.2. Since the population data in UA2012 is based on residential addresses no population is associated with ME_traffic, ME_other and the industrial areas defined as ME_work in Fig. 4a. In contrast, as per definition of ME_home, higher population totals are noted for this microenvironment (Fig. 4a; Refer to Fig. 2a for ME definition). The spatial distribution for the estimated dynamic population is illustrated in Fig. 4b with population totals in each microenvironment. Some distinct differences between the spatial distribution of the static and annual dynamic populations can be clearly seen in Fig. 4c. Decreases in the population can be noted in ME_home while increases are noted for ME_other, and ME_work as we move from a static population in ME_home to a moving population in all MEs.

The percentage contribution of the population in each ME for the dynamic approach is presented in Fig. 5 with 72%, 15%, 4% and 9% in ME_home, ME_other, ME_traffic and ME_work, respectively. Overall, we have estimated that people will mostly be in ME_home both in weekdays and weekends. In addition, we increase the amount of people in ME_other in the weekends and assign a portion of the population to ME_traffic. As no population within the UA2012 data lies within ME_other and ME_traffic, we assign a constant number of people to these MEs depending on the time-activity profiles discussed in Section 2.2.2. Therefore, we do not account for any busy roads or traffic profiles.

The hourly NO_2 concentrations used in this study are obtained from the traffic site of Msida but scaled to better represent urban background concentrations (Refer to Section 2.1). The raw daily NO_2 concentrations measured at Msida as well as the scaled concentrations are illustrated in Fig. 6. Daily NO_2 concentrations at Msida range from 9 μg m^{-3} to 80 μg m^{-3} with the highest concentrations occurring mostly in January. Throughout the rest of the year the range of NO_2 concentrations seems to be largely unaltered. The annual average NO_2 concentrations for 2017 reach 38 μg m^{-3}. The scaled daily NO_2 concentrations used in this study range between 3 and 31 μg m^{-3} with an annual mean of 14 μg m^{-3}. In Europe, naturally occurring background annual NO_2 levels typically range between 0.4 and 9.4 μg m^{-3} while annual urban levels range between 20 and 90 μg m^{-3} with a large range of recorded hourly maxima (EEA, 2019; WHO, 2000).

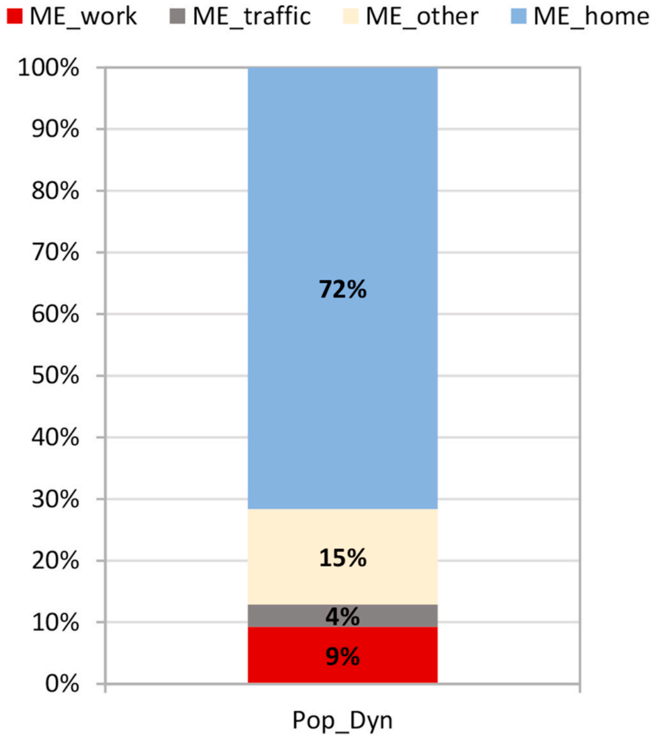


Fig. 5. Percentage contribution of the population in each microenvironment for the annual mean dynamic population estimates (Pop_Dyn).

The annual mean NO_2 total exposure calculated as the annual mean scaled hourly NO_2 concentrations at Msida multiplied by the hourly dynamic population for the Northern Harbour district is illustrated in Fig. 7a. Since the NO_2 concentrations used in this study vary temporally but not spatially, the spatial distribution of the exposure can only be explained by the spatial distribution in the estimated dynamic population (Fig. 4b). Thus, the proportion of the NO_2 exposure in each ME is 72% in ME_home, 15% in ME_other, 9% in ME_work and 4% in ME_traffic. The lowest exposure in all the MEs is found towards the middle of the Northern Harbour district which is the area with the lowest population total and lies within ME_work (Fig. 7a and c). The highest NO_2 exposure is estimated for ME_home (Fig. 7b) as this is the microenvironment with the largest estimated annual mean proportion of the district's population totals (Fig. 5). ME_other which represents any outdoor activity, also includes some high exposure areas (Fig. 7c). This can be explained by the assigned generic diurnal profiles especially for the weekend which include a higher fraction of the population in ME_other. ME_work also exhibits high exposure however, as less time is spent in this microenvironment, the NO_2 exposure is lower compared to ME_home and ME_other (compare Fig. 7b and c to 7e). The NO_2 exposure in ME_traffic is a constant, as a fixed percentage of the population is assigned to this microenvironment for each time step as described previously (Fig. 7d).

In a study focusing on urban population exposure to NO_x emissions in three Baltic Sea harbour cities, Ramacher et al. (2019) estimate similar fractions of exposure in various microenvironments even though their study also included spatially varying pollutant concentrations. The authors suggest up to a 59% contribution to the total NO_2 exposure from ME_home followed by up to 24% in ME_other. ME_work is estimated to constitute between 13 and 19% of the total exposure while the exposure in ME_traffic is estimated to reach a maximum of 9% of the total exposure in all MEs. These similarities are to be expected as the diurnal profiles used in this study are the same as those used in Ramacher et al. (2019).

3.2. *Sensitivity of total NO_2 exposure to indoor infiltration rates and a dynamic population*

In this section we present two sensitivity tests: i) to estimate the total exposure using infiltrated outdoor NO_2 concentrations into ME_home and ME_work as opposed to considering outdoor concentrations for all MEs and ii) to estimate the impact of implementing a static population for every time step as opposed to a more dynamic approach as presented in Section 3.1.

For the first sensitivity test we apply the infiltration factors (IF) described in Section 2.3 to estimate the infiltration of outdoor NO_2 concentrations into indoor MEs while keeping a moving population. The estimated annual mean NO_2 concentrations used when implementing the infiltration factors are illustrated in Fig. 8. Reflecting the infiltration rates used, the lowest annual NO_2 concentrations are estimated for ME_home at 10 μg m^{-3} followed by ME_work at 11 μg m^{-3} and ending with ME_traffic and ME_other having the same unaltered measured outdoor NO_2 concentration at 13 μg m^{-3}. These concentrations generally compare well with other measured indoor NO_2 concentrations. For

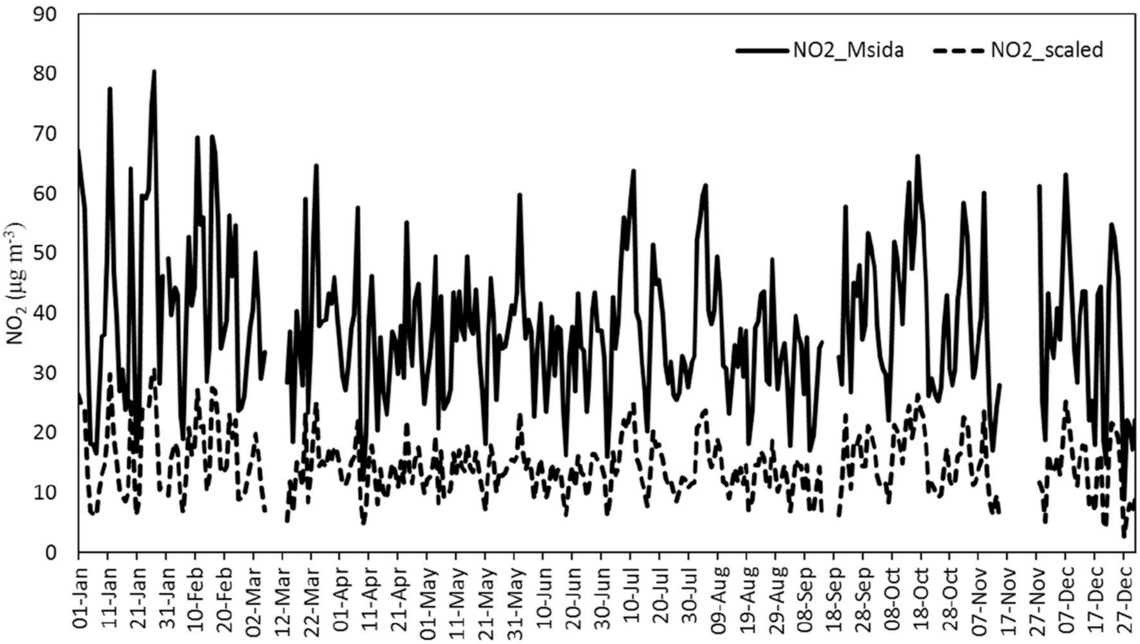


Fig. 6. Daily NO_2 concentrations measured at the traffic site of Msida in 2017 (solid) together with the scaled NO_2 concentrations following the procedure described in Section 2.1 to represent urban background concentrations (dashed) within the district. Missing data was omitted from the analysis.

example the mean indoor NO_2 levels in 34 buildings in Sweden reach 10 μg m^{-3} (Wichmann et al., 2010) while the indoor concentrations in a study conducted in Spain range from 4 μg m^{-3} to 21 μg m^{-3} in rural and urban areas, respectively (Villanueva et al., 2018). This suggests that the estimated indoor NO_2 concentrations in this study are within range of measured European levels. In this section, this test will be referred to as "DynP_IF" and to facilitate the comparison to other sensitivity test we will take this as the reference.

Table 4 represents the total exposure for all the different sensitivity tests together with the set up details. Results suggest that the total exposure estimated using the outdoor NO_2 concentration for all MEs and a moving population (Section 3.1; DynP_noIF) is 25.19% higher compared to DynP_IF. Assuming a static population for all time steps, we find that when the outdoor concentration is used to represent the NO_2 in all MEs the total eposure is 25.19% higher then the reference estimate (StatP_noIF compared to DynP_IF). In contrast the total exposure estimate for the static population with infiltrated NO_2 concentrations for ME_home and ME_work is 3.26% less compared to the DynP_IF estimate (StatP_IF compared to DynP_IF). These results highlight the importance of adequately representing indoor air pollutant concentrations as exposure results from outdoor concentrations may lead to higher exposure estimates by up to 25%. In addition, implementing an estimation of the moving population results in a higher total exposure estimate as opposed to a static population approach by up to 3%. As expected, these results are consistent with results of Ramacher et al. (2019) for Rostock, Riga and Gdansk-Gdynia.

Although results suggest a higher total exposure when assuming outdoor NO_2 concentrations within every ME as opposed to lower concentrations indoors (DynP_noIF compared to DynP_IF), we note that in all our calculations no indoor sources are considered. The presence of indoor sources especially in ME_home in conjunction with infiltrated outdoor NO_2 concentrations can result in a higher indoor exposure compared to the exposure in outdoor MEs. Dimitroulopoulou et al. (2006) suggest an increasing I/O ratio for NO_2 following a decrease in outdoor concentrations as indoor sources contribute more compared to the infiltration from outdoors. In addition, other characteristics can influence indoor NO_2 concentrations. For example in offices and classrooms, the type of ventilation (natural or mechanical) and different air exchange rates (Challoner and Gill, 2014; Wichmann et al., 2010), furnishings and cooking methods (WHO, 2010) as well as proximity to outdoor sources (Al-Hemoud et al., 2017; Villanueva et al., 2018) have all been found to influence indoor NO_2 concentrations.

The proportion of the exposure within each microenvironment for the different sensitivity tests described in Table 4 is illustrated in Fig. 9. Implementing a moving population approach results in an exposure that is highest in ME_home followed by ME_other, ME_work and ME_traffic, respectively. This is the same distribution as that noted for the dynamic population totals in Fig. 5. When considering infiltration rates in conjuction with a dynamic population (DynP_IF), the distribution changes compared to that in DynP_noIF. The estimated NO_2 exposure for DynP_IF in ME_home is 5% less compared to DyP_noIF as a result of lower NO_2 concentrations in this ME. In addition, the proportion of the NO_2 exposure in ME_other and ME_traffic increases by 3% and 1%, respectively while no changes are noted for ME_work. Using a dynamic population we highlight ME_home as the microenvironment with the highest NO_2 exposure followed by ME_other. This exposure is a conservative one as no indoor sources are taken into account. Thus targeted policies to reduce NO_2 exposure should focus primarily on this ME.

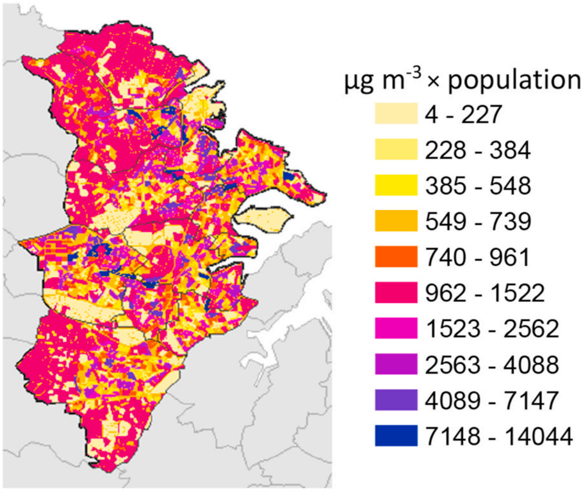
4. Summary and conclusions

This study is the first to present an estimate of the total NO_2 population exposure in a polluted and densely populated district in Malta. To estimate the total NO_2 exposure we use a generic method to simulate a moving population together with outdoor NO_2 concentrations from a traffic site in Msida that lies within the Northern Harbour district.

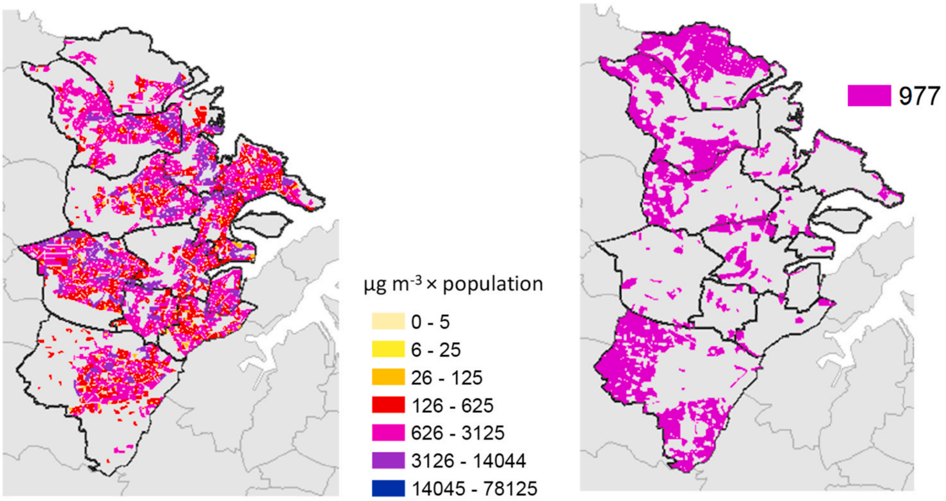
Results highlight differences between a static population distribution which is typically used in exposure assessments and a moving population. Following the definitions of the different microenvironments we estimate that the greatest portion of the population is mostly in ME_home followed by ME_other, ME_work and ME_traffic, respectively when implementing a dynamic population approach. The total exposure is then estimated across all MEs. The spatial distribution of the annual total NO_2 exposure is generally the same as the dynamical population distribution as the NO_2 concentrations are constant for all MEs and only vary temporally.

In a sensitivity test we estimate the indoor NO_2 concentrations in the indoor MEs by using infiltrations factors established in literature. The

(a) ME_total



(b) ME_home (c) ME_other



(d) ME_traffic (e) ME_work

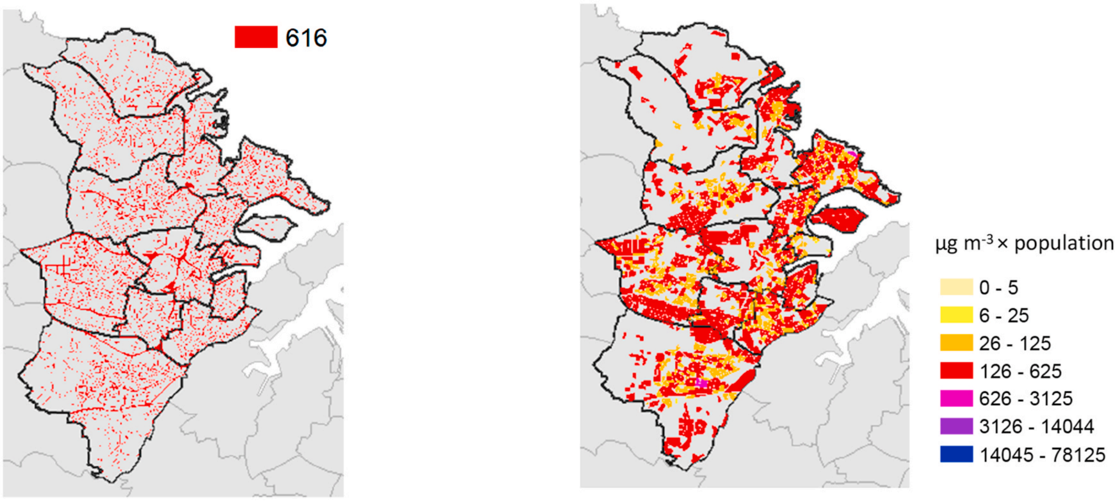


Fig. 7. The NO_2 exposure calculated as the annual mean of the hourly dynamic population multiplied by the hourly NO_2 scaled concentrations at the traffic site of Msida (µg m^{-3} × population) for (a) all MEs, (b) ME_home, (c) ME_other, (d) ME_traffic and (e) ME_work. N.B. colour scale for (a) is different compared to (b–e). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

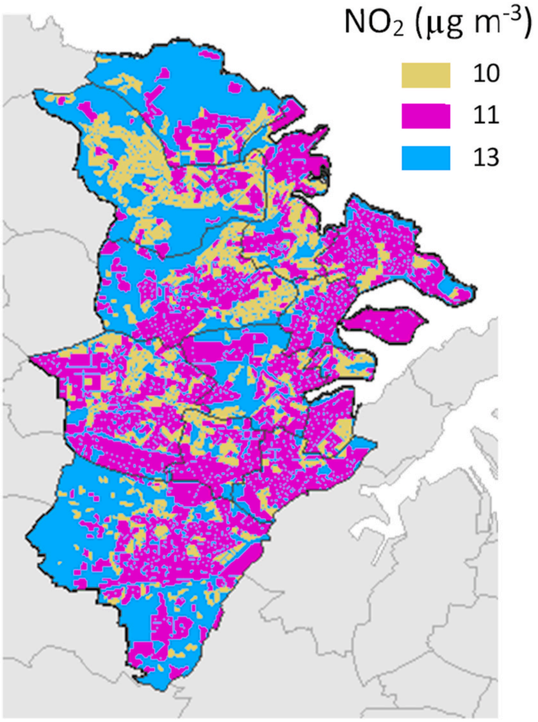


Fig. 8. Annual mean NO_2 concentrations including summer and winter infiltration rates for the indoor microenvironments: ME_home and ME_work.

Table 4
Details of the different sensitivity test together with the percentage difference in each test compared to the reference.

Test Name	Population	IF	Total Exposure (μg m^{-3} x pop.)	% Diff (compared to Ref.)
DynP_IF (Ref.)	Dynamic	Yes	1.6×10^6	/
DynP_noIF	Dynamic	No	2.0×10^6	+25.19%
StatP_IF	Static	Yes	1.6×10^6	−3.26%
StatP_noIF	Static	No	2.0×10^6	+25.19%

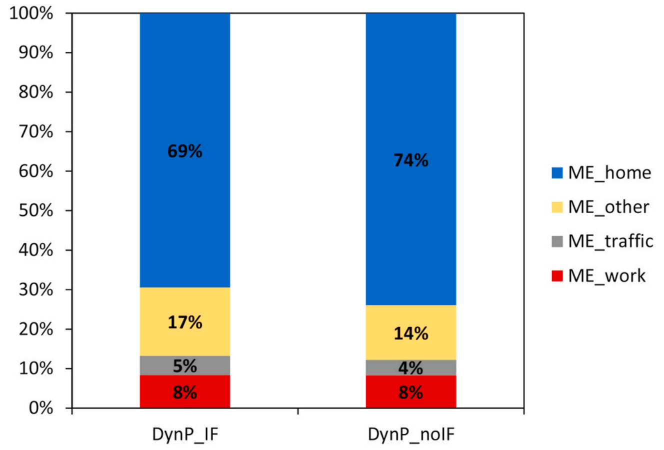


Fig. 9. Percentage contribution of the exposure in each microenvironment for each of the sensitivity test described in Table 4.

estimated total exposure using outdoor NO_2 concentrations for all MEs is 25% higher compared to the estimated total exposure with varying indoor and outdoor NO_2 concentrations. The total exposure is also affected if a static population is assumed instead of a dynamic one with results for a static population being 3% less compared to a more dynamic approach. These results highlight the importance of adequately representing NO_2 concentrations in the different microenvironments as well as the importance of considering population mobility as these will alter the estimated exposure substantially.

A limitation of this study is the use of NO_2 concentrations at a fixed site to represent the NO_2 exposure throughout the Northern Harbour district with scaling to represent urban background concentrations. Consequently, exposure estimates are somewhat conservative as high levels of pollution exposure for MEs close to major traffic routes are not accounted for. Future work should focus on developing new tools to better represent the air pollution gradients typical of urban and densely populated areas such as the Northern Harbour district. Using outdoor NO_2 concentrations as a proxy for total exposure as well as infiltration factors to simulate infiltrated pollutant concentrations is a method widely used in literature however, in this study no indoor sources are considered. Future research would benefit from a more detailed representation of the local indoor exposures. Personal air pollution monitoring in conjunction with GPS technology would give the exact pollutant exposure at high temporal and spatial resolutions however, this is costly, time consuming and subject specific. Nonetheless, other methods can be used to better represent the spatial distribution of both outdoor and indoor data. For example, the use of chemistry transport models (which are still under development for our country) as well as machine learning methods to simulate outdoor concentrations (e.g. Gariazzo et al., 2020) and indoor probabilistic and/or statistical models to estimate indoor concentrations using established I/O ratios under different scenarios (e.g. Dimitroulopoulou et al., 2006; Li and Friedrich, 2019; Terry et al., 2014). In addition, in this study we have assigned a constant number of people to ME_traffic for every time step. A better representation of the population mobility within this ME is required to simulate for example the number of people stuck in traffic in major roads during rush hours.

Nonetheless this study presents estimates for indoor NO_2 concentrations (of outdoor origin) in the Northern Harbour district of Malta together with the corresponding NO_2 exposure in the different MEs using both a static and a dynamic population approach. The results presented highlight the importance for local future research to focus on the indoor microenvironments especially ME_home as the NO_2 exposure is highest in this ME. Exposure assessments such as that presented in this study are essential to aid the development of targeted policies to limit such exposures.

CRediT authorship contribution statement

Sara Fenech: Conceptualization, Methodology, Software, and, Formal analysis, Writing - original draft, preparation. **Noel J. Aquilina:** Conceptualization, Methodology, Writing - review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.atmosenv.2020.117918.

References

Al-Hemoud, A., Al-Awadi, L., Al-Rashidi, M., Abdul, K., Al-khayat, A., Behbehani, W., 2017. Comparison of indoor air quality in schools: urban vs. Industrial "oil & gas" zones in Kuwait. Build. Environ. 122, 50–60. https://doi.org/10.1016/j.buildenv.2017.06.001.

Borrego, C., Sa, E., Monteiro, A., Ferreira, J., Miranda, A.I., 2009. Forecasting human exposure to atmospheric pollutants in Portugal – a modelling approach. Atmos. Environ. 43 (36), 5796–5806. https://doi.org/10.1016/j.atmosenv.2009.07.049.

Challoner, A., Gill, L., 2014. Indoor/outdoor air pollution relationships in ten commercial buildings: PM2.5 and NO2. Build. Environ. 80, 159–173. https://doi.org/10.1016/j.buildenv.2014.05.032.

COMEAP, 2015. Interim statement on quantifying the association of long-term average concentrations of nitrogen dioxide and mortality [online] Available from: https://www.gov.uk/government/uploads/system/uploads/attachment_data/file/485373/COMEAP_NO2_Mortality_Interim_Statement.pdf.

COMEAP, 2018. Associations of Long-Term Average Concentrations of Nitrogen Dioxide with Mortality.

Copernicus Land Monitoring Service, 2012a. Mapping guide v4.7 for a European urban Atlas [online] Available from: https://land.copernicus.eu/local/urban-atlas/urban-atlas-2012?tab=metadata.

Copernicus Land Monitoring Service, 2012. Urban Atlas 2012 [online] Available from: https://land.copernicus.eu/local/urban-atlas/urban-atlas-2012. Accessed 30 April 2020b.

Crouse, D.L., Peters, P.A., Hystad, P., Brook, J.R., van Donkelaar, A., Martin, R.V., Villeneuve, P.J., Jerrett, M., Goldberg, M.S., Arden Pope, C., Brauer, M., Brook, R.D., Robichaud, A., Menard, R., Burnett, R.T., 2015. Ambient PM2.5, O3, and NO2exposures and associations with mortality over 16 years of follow-up in the canadian census health and environment cohort (CanCHEC). Environ. Health Perspect. 123 (11), 1180–1186. https://doi.org/10.1289/ehp.1409276.

Dimitroulopoulou, C., Ashmore, M.R., Hill, M.T.R., Byrne, M.A., Kinnersley, R., 2006. INDAIR : a probabilistic model of indoor air pollution in UK homes. Atmos. Evironment 40, 6362–6379. https://doi.org/10.1016/j.atmosenv.2006.05.047.

Dionisio, K.L., Baxter, L.K., Burke, J., Özkaynak, H., 2016. The importance of the exposure metric in air pollution epidemiology studies : when does it matter , and why ? Air Qual. Atmos. Heal. 495–502. https://doi.org/10.1007/s11869-015-0356-1.

EEA, 2019. Air Quality in Europe — 2019 Report. Luxembourg.

Environment Resource Authority (ERA), 2018. Chapter 2 : Ambient Air Reporting Status from 2009 to 2015.

Fenech, S., Aquilina, N.J., 2020. Trends in ambient ozone , nitrogen dioxide , and particulate matter concentrations over the Maltese Islands and the corresponding health impacts. Sci. Total Environ. 700, 134527. https://doi.org/10.1016/j.scitotenv.2019.134527.

Gaffin, J.M., Hauptman, M., Petty, C.R., Sheehan, W.J., Lai, P.S., Wolfson, J.M., Gold, D. R., Coull, B.A., 2018. Nitrogen dioxide exposure in school classrooms of inner-city children with asthma. J. Allergy Clin. Immunol. 141 (6) https://doi.org/10.1016/j.jaci.2017.08.028.

Gariazzo, C., Carlino, G., Silibello, C., Renzi, M., Finardi, S., Pepe, N., Radice, P., Forastiere, F., Michelozzi, P., Viegi, G., Stafoggia, M., 2020. A multi-city air pollution population exposure study : Combined use of chemical-transport and random-Forest models with dynamic population data. Sci. Total Environ. 724, 138102. https://doi.org/10.1016/j.scitotenv.2020.138102.

Guarnieri, M., Balmes, J.R., 2014. Outdoor air pollution and asthma. Lancet 383 (9928), 1581–1592. https://doi.org/10.1016/S0140-6736(14)60617-6.Outdoor.

Guerreiro, C.B.B., Foltescu, V., de Leeuw, F., 2014. Air quality status and trends in Europe. Atmos. Environ. 98, 376–384. https://doi.org/10.1016/j.atmosenv.2014.09.017.

He, B., Huang, J.V., Ki, M., Lun, S., Yeung, A., Ling, L., Li, A.M., Leung, G.M., Schooling, C.M., 2019. The association of early-life exposure to air pollution with lung function at $\sim$ 17 . 5 years in the " Children of 1997 " Hong Kong Chinese Birth Cohort. Environ. Int. 123, 444–450. https://doi.org/10.1016/j.envint.2018.11.073. November 2018.

Hicklin, W., Farrugia, P.S., Sinagra, E., 2018. Investigations of VOCs in and around buildings close to service stations. Atmos. Environ. 172, 93–101. https://doi.org/10.1016/j.atmosenv.2017.10.012.

Kazakos, V., Luo, Z., Ewart, I., 2020. Quantifying the health burden misclassification from the use of di ff erent pm 2 . 5 exposure tier Models : a case study of London. Int. J. Environ. Res. Publ. Health. https://doi.org/10.3390/ijerph17031099.

Krewski, D., Jerrett, M., Burnett, R.T., Ma, R., Hughes, E., 2009. Extended Follow-Up and Spatial Analysis of the American Cancer Society Study Linking Particulate Air Pollution and Mortality. Boston, Massachusetts.

Li, N., Friedrich, R., 2019. Methodology for estimating the lifelong exposure to PM2.5 and NO2 - the application to European population subgroups. Atmosphere. https://doi.org/10.3390/atmos10090507.

Malley, C.S., Henze, D.K., Kuylenstierna, J.C.I., Vallack, H.W., Davila, Y., Anenberg, S.C., Turner, M.C., Ashmore, M.R., 2017. Updated global estimates of respiratory mortality in adults $\geq$ 30 years of age attributable to long-term ozone exposure. Environ. Health Perspect. 125 (8), 1–9. https://doi.org/10.1289/EHP1390.

McGrath, J.A., Sheahan, J.N., Dimitroulopoulou, C., Ashmore, M.R., Terry, A.C., Byrne, M.A., 2017. PM exposure variations due to different time activity profile simulations within a single dwelling. Build. Environ. 116, 55–63. https://doi.org/10.1016/j.buildenv.2017.02.002.

Monks, P.S., Archibald, A.T., Colette, A., Cooper, O., Coyle, M., Derwent, R., Fowler, D., Granier, C., Law, K.S., Mills, G.E., Stevenson, D.S., Tarasova, O., Thouret, V., Von Schneidemesser, E., Sommariva, R., Wild, O., Williams, M.L., 2015. Tropospheric ozone and its precursors from the urban to the global scale from air quality to short-lived climate forcer. Atmos. Chem. Phys. 15 (15), 8889–8973. https://doi.org/10.5194/acp-15-8889-2015.

National Statistics Office Malta (NSO), 2017. Regional Statistics Malta.

NSO, 2014. Census of Population and Housing 2011.

NSO, 2019. Regional Statistics Malta 2019 Edition. Valletta.

Özkaynak, H., Baxter, L.K., Dionisio, K.L., Burke, J., 2013. Air pollution exposure prediction approaches used in air pollution epidemiology studies. J. Expo. Sci. Environ. Epidemiol. https://doi.org/10.1038/jes.2013.15.

Pegas, P.N., Alves, C.A., Evtyugina, M.G., Nunes, T., Cerqueira, M., Franchi, M., Pio, C.A., Almeida, S.M., Freitas, M.C., 2011. Indoor Air Quality in Elementary Schools of Lisbon in Spring, pp. 455–468. https://doi.org/10.1007/s10653-010-9345-3.

Ramacher, M.O.P., Karl, M., 2020. Integrating modes of transport in a dynamic modelling approach to evaluate population exposure to ambient NO2 and PM2.5 pollution in urban areas. Int. J. Environ. Res. Publ. Health 1–35.

Ramacher, M.O.P., Karl, M., Bieser, J., Jalkanen, J., Johansson, L., 2019. Urban population exposure to NO x emissions from local shipping in three Baltic Sea harbour cities – a generic approach. Atmos. Chem. Phys. 9153–9179. https://doi.org/10.5194/acp-19-9153-2019.

RCP, 2016. Every Breath We Take the Lifelong Impact of Air Pollution. London.

Reis, S., Vieno, M., Carnell, E.J., Beck, R., Clemens, T., Dragosits, U., Tomlinson, S.J., Leaver, D., Heal, M.R., 2018. The influence of residential and workday population mobility on exposure to air pollution in the UK. Environ. Int. 121 (October), 803–813. https://doi.org/10.1016/j.envint.2018.10.005.

Salonen, H., Salthammer, T., Morawska, L., 2019. Human exposure to NO 2 in school and office indoor environments. Environ. Int. 130 (May) https://doi.org/10.1016/j.envint.2019.05.081.

Seinfeld, J.H., Pandis, S.N., 2016. Atmospheric Chemistry and Physics: from Air Pollution to Climate Change, third ed. John Wiley & Sons, Inc [online] Available from: https://books.google.com.mt/books?id=n_RmCgAAQBAJ&lpg=PR5&ots=gRNeDNocdI&lr&pg=PR10#v=onepage&q&f=false.

Terry, A.C., Carslaw, N., Ashmore, M., Dimitroulopoulou, S., Carslaw, D.C., 2014. Occupant exposure to indoor air pollutants in modern European offices : an integrated modelling approach. Atmos. Environ. 82, 9–16. https://doi.org/10.1016/j.atmosenv.2013.09.042.

Turner, M.C., Jerrett, M., Pope, C.A., Krewski, D., Gapstur, S.M., Diver, W.R., Beckerman, B.S., Marshall, J.D., Su, J., Crouse, D.L., Burnett, R.T., 2015. Long-term ozone exposure and mortality in a large prospective study. Am. J. Respir. Crit. Care Med. 193 (10), 1134–1142. https://doi.org/10.1164/rccm.201508-1633OC.

US Environmental Protection Agency (EPA), 2017. Our nation's air [online] Available from: https://gispub.epa.gov/air/trendsreport/2017/.

Vella, R., 2019. A Study on the Dynamics of Particulate Matter Infiltration in Buildings. University of Malta.

Villanueva, F., Tapia, A., Lara, S., Amo-Salas, M., 2018. Indoor and outdoor air concentrations of volatile organic compounds and NO2 in schools of urban, industrial and rural areas in Central-Southern Spain. Sci. Total Environ. 622–623, 222–235. https://doi.org/10.1016/j.scitotenv.2017.11.274.

WHO, 2000. Nitrogen dioxide. In: Air Quality Guidelines -, , second ed.vol. 3, pp. 1–33 Denmark.

WHO, 2005. Air Quality Guidelines. Global Update 2005. Copenhagen.

WHO, 2010. WHO guidelines for indoor air quality [online] Available from: www.euro.who.int/_data/assets/pdf_file/0009/128169/e94535.pdf.

WHO, 2013. Health Risks of Air Pollution in Europe – HRAPIE project: Recommendations for concentration-response functions for cost-benefit analysis of particulate matter, ozone and nitrogen dioxide [online] Available from: http://www.euro.who.int/_data/assets/pdf_file/0006/238956/Health_risks_air_pollution_HRAPIE_project.pdf?ua=1.

WHO, 2015. Review of Evidence on Health Aspects of Air Pollution. REVIHAAP project.

Wichmann, J., Lind, T., Nilsson, M.A., Bellander, T., 2010. PM2.5 , soot and NO2 indoor-outdoor relationships at homes, pre-schools and schools in Stockholm, Sweden, Atmos. Environ. Times 44 (2), 4536–4544. https://doi.org/10.1016/j.atmosenv.2010.08.023.

Worldometer. Malta demographics [online] Available from: https://www.worldometers.info/demographics/malta-demographics/. (Accessed 12 May 2020), 2020.