



## Potential ambient NO<sub>2</sub> abatement by applying photocatalytic materials in a Spanish city and analysis of short-term effect on human mortality<sup>☆</sup>

Jaime Fernández-Pampillón<sup>a,b</sup>, Magdalena Palacios<sup>a</sup>, Lourdes Núñez<sup>a,\*</sup>, Manuel Pujadas<sup>a</sup>, Begoña Artíñano<sup>a</sup>

<sup>a</sup> Research Centre for Energy, Environment and Technology (CIEMAT), Madrid, 28040, Spain

<sup>b</sup> The National Distance Education University (UNED), Madrid, 28232, Spain

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### ABSTRACT

Road traffic is the main contributor to NO<sub>2</sub> emissions in many European cities, causing that the current limit values for the protection of human health are exceeded. The use of photocatalytic compounds that incorporate titanium dioxide (TiO<sub>2</sub>) is frequently proposed as abatement technology but its depolluting effectiveness on a real scale is still being investigated.

In this work, the potential removal capacity of NO<sub>2</sub> that selected TiO<sub>2</sub>-based materials would have if they were implemented in a street in the municipality of Alcobendas (Community of Madrid, Spain) has been evaluated. The number of avoided NO<sub>2</sub>-related deaths over the locality across the period 2001–2019 have been inferred. Moreover, the saving associated with the estimated removal of ambient NO<sub>2</sub> due to the use of photocatalytic materials and costs generated by their acquisition and implementation in the selected urban environment were briefly studied.

Attributable mortality due to NO<sub>2</sub> concentrations for Alcobendas has been estimated in 289 deaths, being 9241 the total deaths due to natural cause. This presents a monthly variation associated with the evolution of both mortality due to natural causes and the average concentrations of NO<sub>2</sub>.

The reduction in mortality via the hypothetical implantation of photocatalytic materials throughout the municipality, assuming ideal conditions for their optimal performance, would be a maximum of 3%.

In addition, a saving of €5708 yr<sup>-1</sup> km<sup>-2</sup> related to NO<sub>x</sub> damage costs of transport was obtained. A total cost of k€4750.5 km<sup>-2</sup> was associated to the purchase of photocatalytic materials and their application to all surfaces in that area.

This technology has a big elimination potential in controlled conditions but a low reduction of ambient NO<sub>2</sub> is provided when implemented in real outdoor urban scenarios. Its use can be recommended incorporated into engineering designs and applications, complementing other abatement measures, to reduce NO<sub>2</sub> mortality in urban areas.

### 1. Introduction

Air pollution is one of the biggest environmental health risk factors worldwide and increases the whole diseases burden (World Health Organization, 2016), (Health Effects Institute, 2020), (European Environment Agency, 2021). Global assessments of ambient air pollution suggest between 4 and 9 million deaths annually (World Health Organization, 2021), (Murray et al., 2020).

Nitrogen dioxide (NO<sub>2</sub>) is an air pollutant for which the evidence of

short-term health effects is increasing. Several studies point a causal relationship between short-term NO<sub>2</sub> exposure and respiratory effects and with cardiovascular effects and total mortality (European Environment Agency, 2021), (Environmental Protection Agency, 2016), (World Health Organization, 2013a), (Chen et al., 2018), (Linares et al., 2018). Additionally, there is some evidence supporting a role for long-term NO<sub>2</sub> in increasing all non-accidental and, especially, respiratory mortality (World Health Organization, 2021) or even in causing cardiovascular mortality (Schneider et al., 2018).

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\* Corresponding author.

E-mail address: [lourdes.nunez@ciemat.es](mailto:lourdes.nunez@ciemat.es) (L. Núñez).

The road transport sector is the principal source of nitrogen oxides ( $\text{NO}_x = \text{NO} + \text{NO}_2$ ) emissions (39% in the EU-27 in 2019) where high  $\text{NO}_2$  concentrations above the annual limit value (Directive, 2008/30/EC) are frequently observed specially at traffic stations located in densely populated urban environments. Even though during the last decades a slight decrease in the annual means of  $\text{NO}_x$  pollution in European cities has become evident,  $\text{NO}_2$  concentrations remain and are still a problem to be solved (European Environment Agency, 2021), (Municipality of Madrid, 2017). The reason is twofold. On the one hand, the ratio of  $\text{NO}_2/\text{NO}_x$  emissions from road traffic has increased (mainly due to the introduction of certain types of diesel particulate filters in buses and the high penetration of diesel passenger cars in the market) (Carslaw et al., 2007). On the other hand, secondary  $\text{NO}_2$ , formed from directly emitted nitric oxide (NO) and, partially, ozone ( $\text{O}_3$ ) or peroxide radicals ( $\text{RO}_2$ ), does not decrease substantially due to the strongly non-linear dependence of  $\text{NO}_x$  levels with the already mentioned reactions and the photolysis of  $\text{NO}_2$  (Leighton balance) (Palacios et al., 2002), (Kurtenbach et al., 2012). This makes it difficult to reduce ambient  $\text{NO}_2$ .

In particular, road traffic is the main source of anthropogenic  $\text{NO}_x$  emissions in the Madrid region (57%) (Palacios et al., 2001) and 78% of local sources concerning  $\text{NO}_2$  annual average concentration in the metropolitan urban area (Borge et al., 2014). Both the city and other agglomerations of the region frequently exceed the hourly and annual limits for  $\text{NO}_2$  (mainly from October to February) (Ministry of Agriculture and Fishing and Food and Environment, 2017). The Madrid region, located in the centre of the Iberian Peninsula, is frequently under the influence of high pressures that cause poor atmospheric ventilation and favour significant episodes of air pollution, especially in winter and, in general, with moderate ambient concentrations of  $\text{NO}_2$  and particulate matter (Martín et al., 2001), (Pujadas et al., 2000). On the other hand, the advection of polluted air masses from the Madrid metropolitan area, which are added to the local emissions of certain municipalities, complicate the problem of improving air quality in some areas near Madrid city (for example, the North agglomeration) (Community of Madrid, 2021).

The need to decrease the contribution of  $\text{NO}_x$  to air pollution in urban areas has favoured the introduction and evaluation of different mitigation strategies. In addition to the measures adopted to reduce  $\text{NO}_x$  emissions, it is necessary to implement other control measures that eliminate the  $\text{NO}_2$  present in the air (Vedrenne et al., 2015). Among the available air pollution control options with potential success in eliminating ambient  $\text{NO}_x$ , it is worth highlighting the use of construction materials that incorporate photocatalyst compounds such as titanium dioxide ( $\text{TiO}_2$ ). It has been proved in numerous laboratory tests that this kind of materials, activated by sunlight, allow the elimination of those pollutants by heterogeneous photocatalysis (Mendoza et al., 2015), (Martinez et al., 2011), (Ballari et al., 2010), (Laufs et al., 2010), (Sikkema et al., 2015), (de Melo and Trichês, 2012), (Zouzelka and Rathousky, 2017), (Ángelo et al., 2014), (Mothes et al., 2018), (Bengtsson and Castellote, 2010), (Karapati et al., 2014). In recent decades, the development of this technology has led to the commercialization of different building materials that have found a field of application for ambient air depollution in European urban environments (Maggos et al., 2008), (Ballari and Brouwers, 2013), (Guerrini and Peccati, 2007), (Moussiopoulos et al., 2008), (Boonen and Beeldens, 2014), (Chen and Chu, 2011), (IPL, 2010), (TERA environment, 2009), (Gallus et al., 2015a), (Gallus et al., 2015b), (Barratt et al., 2012), (Cordero et al., 2021).

Heterogeneous photocatalysis applied to air pollution mitigation implies the interaction between a solid catalyst and the gas phase. The pollutant is transferred to and adsorbed on the photoactive surface  $\text{TiO}_2$  centres; next, the absorption of UV-A radiation by  $\text{TiO}_2$  promotes the formation of electron/hole pairs; and lastly, adsorbed  $\text{O}_2$  and  $\text{H}_2\text{O}$  reacts to form reactive oxygen species that oxidize the nitrogen oxides and the nitrate produced can be finally desorbed. Thus, photocatalytic activity,

described in further detail elsewhere, is presented in Fig. 1 (adapted from (Monge et al., 2010), (Dillert et al., 2013), (Ballari et al., 2011), (Herrmann, 2010)).

The latest trends in this technology are aimed at improving performance through greater control of the reaction through new additives, as well as the introduction of different mixtures of photocatalysts. Also, the possibility of extending the useful spectral range from UVA to visible is being investigated (Ochiai and Fujishima, 2012), (Ahmad et al., 2016). On the other hand, research on the kinetics characteristic of the different chemical processes that take place continues, enormously dependent on the substrate to which  $\text{TiO}_2$  is incorporated and the additives that are used (Mothes et al., 2018), (Ángelo et al., 2013), (Mills et al., 2016), (Mothes et al., 2016). All this draws an enormous field of experimentation and applicability, referred both to applications in outdoor (urban environment, fundamentally) and indoor. A great piece of information is continually emerging as far as the market launch of new building photocatalytic materials with big potential to reduce  $\text{NO}_x$  ambient concentrations. These announcements are based on promising laboratory test under controlled conditions but their efficiency to eliminate these pollutants on a real scale is plenty of uncertainties and the possible positive impact in the air quality at real urban areas has to be carefully studied (Air Quality Expert Group of the Department for Environment Food and Rural Affairs, 2016).

Recently, the LIFE MINOX-STREET demonstration project, co-funded by the EU, has had as its first objective to provide evidence from rigorous tests and trials on the physical-chemical properties and the expected efficiency of several commercial photocatalytic materials. Secondly, the depolluting capabilities of the three most promising photoactive products selected to be implemented in three different urban surfaces (road, sidewalk and façade, respectively) in the municipality of Alcobendas (located 10 km northwest of the city of Madrid) were evaluated at real scale (Palacios et al., 2018), (European Commission, 2023).

Among the objectives achieved for this project, a characterisation of the sink effect on  $\text{NO}_x$  levels on different photocatalytic surfaces under controlled ambient conditions was performed (Palacios et al., 2015a, 2015b, 2015c, 2015d, 2015e). This allowed to analyse the potential combined efficiency of several photocatalytic materials when are applied to different elements of the urban environment at street level to improve air quality.

The main contribution of this work refers to the methodology presented to evaluate the capacity of the use of photocatalytic materials to reduce ambient  $\text{NO}_2$  concentration in a city of the Community of Madrid and, subsequently, to estimate the decrease in short-term natural cause mortality due to nitrogen dioxide if this technology would have been used during the last two decades, as well as the costs derived from its implementation. Finally, the possible potential benefit has been estimated, in terms of mortality reduction, if the air quality objectives proposed by WHO were achieved.

## 2. Methodology

First of all, maximum reduction in  $\text{NO}_2$  ambient concentration potentially expected as a consequence of the implementation of photocatalytic materials was estimated. The methodology utilized has been based on the different results obtained in the LIFE MINOX-STREET project.

Following short-term association between  $\text{NO}_2$  concentrations and natural cause mortality, named as relative risk, referred to the inhabitants of the municipality of Alcobendas during the period 2001–2019, has been used to compute attributable mortality due to  $\text{NO}_2$  concentrations in three different scenarios. The first one is considering the city with its actual conditions, the second one is an ideal situation where all roads, sidewalks and facades are treated with photocatalytic coatings and, in the third one, the  $\text{NO}_2$  ambient concentration is  $20 \mu\text{g m}^{-3}$ , the AQG level proposed as interim target 3 for annual mean  $\text{NO}_2$  concentration by WHO (World Health Organization, 2021), (World

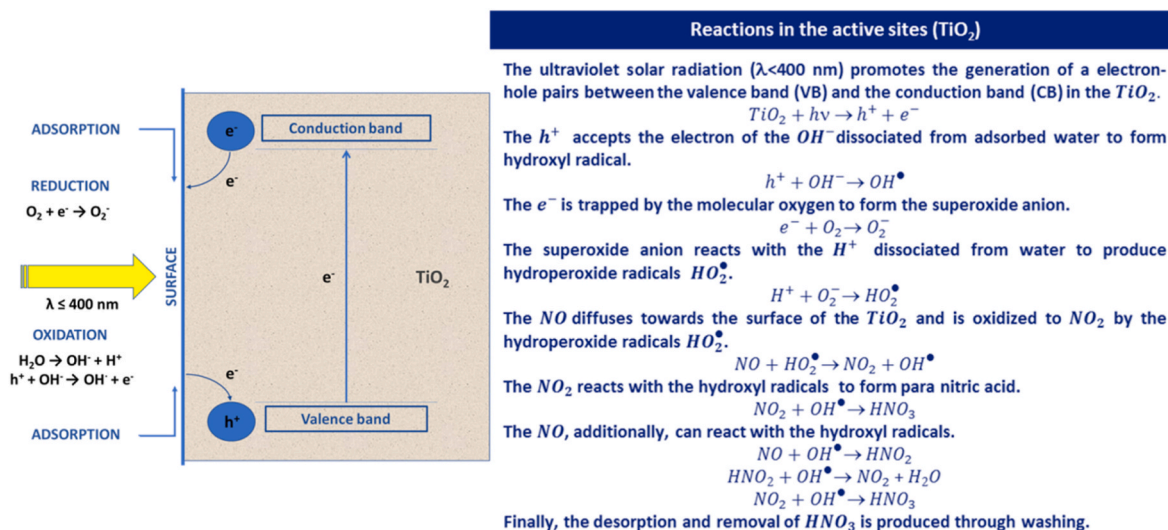


Fig. 1. Photocatalytic reaction mechanism of nitrogen oxides on  $TiO_2$  (adapted from (Monge et al., 2010), (Dillert et al., 2013), (Ballari et al., 2011), (Herrmann, 2010)).

Health Organization, 2013b).

Subsequently, the benefit in terms of mortality that the application of photocatalytic materials would have throughout the city was estimated, as well as what would have been the impact associated with a reduction in the concentration of  $NO_2$  adjusted to certain regulated limits. Furthermore, a rough study of the associated costs saved from removing ambient  $NO_2$  and those derived from the acquisition and arrangement of the materials in the urban environment selected was undertaken.

Such methodology has been applied to Alcobendas, which currently has a population of around 117000 inhabitants (Statistics National Institute, 2023), well below the great European metropolises but similar to many other medium-sized municipalities located in the Community of Madrid. Beyond the prevailing air pollution conditions and regardless of the impact that the reduction of air pollution by  $NO_2$  produces on the number of deaths in this specific location, the methodology presented is applicable to similar studies at any city.

2.1. Ambient  $NO_2$  reduction due to the implementation of photocatalytic materials

Even when no available measurements could be used to calculate  $NO_2$  depolluting efficiency, laboratory assays under the ISO 22197-1:2007 international standard method (ISO, 2007) were used to investigate the  $NO$  depolluting performance of a variety of commercial photocatalytic materials and select three coatings that finally were implemented in Alcobendas (a water emulsion on a bituminous concrete pavement, a photocatalytic coating on a concrete pavement and a photocatalytic mono-component paint on a brick-facade), because their notable  $NO$  purifying efficiency. Additionally, these data were used to estimate the corresponding  $NO$  surface deposition velocities by following a first-order kinetic approximation as it is described below (Palacios et al., 2015a, 2015d) and to approximate  $NO_2$  surface deposition velocities needed to compute ambient  $NO_2$  reduction due to the implementation of photocatalytic materials in that city.

Thus, a rate coefficient ( $k_r, s^{-1}$ ), can be determined from the experimental data:

$$k_r = - \frac{\ln\left(\frac{C_{out}}{C_{in}}\right)}{t_r} \tag{1}$$

where  $C_{in}$  and  $C_{out}$  (ppbv) are  $NO$  concentration at the inlet and exit of the photo-reactor and  $t_r$  (s) is the reaction time of the gas-phase  $NO$  and the sample.  $k_r$  is directly related to the activity of the sample, on one

hand, but also dependent on the geometry and size of the reactor. Therefore, the dimensionless reactive uptake coefficient,  $\gamma$ , has been introduced in heterogeneous chemistry. The reactive uptake coefficient is defined as the ratio of the number of collisions that lead to reaction over all collisions of the gas-phase reactant with a reactive surface. It can be calculated from  $k_r$  as:

$$\gamma = \frac{4 \cdot k_r}{\bar{v} \cdot S/V} \tag{2}$$

where  $S/V$  ( $m^{-1}$ ) is the ratio of photoactive surface to air volume above this surface and  $\bar{v}$  ( $m s^{-1}$ ) is the mean molecular velocity of the gas  $NO$  defined by the kinetic gas theory:

$$\bar{v} = \sqrt{\frac{8 \cdot R \cdot T}{\pi \cdot M}} \tag{3}$$

where  $R$  ( $8.314 J mol^{-1} K^{-1}$ ) is the ideal gas constant,  $T$  (K) is the absolute temperature and  $M$  ( $kg mol^{-1}$ ) is the molecular mass of  $NO$ . Then, an analogue surface deposition velocity  $V_{surf}$  ( $m s^{-1}$ ) can be defined:

$$V_{surf} = \frac{\gamma \cdot \bar{v}}{4} \tag{4}$$

which can be useful to estimate surface deposition fluxes in atmospheric models through a surface resistance computed from the inverse of the surface deposition velocity. Further details on kinetic parameters are given elsewhere (Ifang et al., 2014).

On the other hand,  $NO_2$  surface deposition velocities for several  $TiO_2$ -based photocatalytic materials, calculated by means of laboratory tests, have given similar values or even one order of magnitude lower than that obtained for  $NO$  (Laufs et al., 2010), (Mothes et al., 2018), (Gallus et al., 2015b), (Air Quality Expert Group of the Department for Environment Food and Rural Affairs, 2016), (Ifang et al., 2014), (Engel et al., 2015). Here,  $NO$  and  $NO_2$  surface deposition velocities are assumed to have the same value for every one of the photoactive materials under consideration in order to evaluate an upper limit of photocatalytic  $NO_2$  degradation and its impact in the surrounding air of a selected stretch of the Paseo de la Chopera avenue (Alcobendas). For this purpose, it is also presumed that  $NO_2$  photocatalytic decomposition follows a first order kinetics in the street under specific atmospheric conditions, reproducing a plugged flow reactor situation in which the air mass travels longitudinal through the canyon without dilution to the upper atmosphere. Then, assessing the  $NO_2$  potential remediation ( $(C_{out}-C_{in})/C_{in}$ ) for the study street is possible by using above

expressions (1–4). Finally, this maximum NO<sub>2</sub> removal value was considered to be the same for the whole municipality of Alcobendas.

## 2.2. Attributable mortality due to NO<sub>2</sub> in real urban scenario

In order to compute the number of deaths attributable to NO<sub>2</sub> concentrations for the municipality of Alcobendas in the whole period considered, the methodology presented by Tobías et al. has been followed (Tobías et al., 2015). Previous studies indicate that in the case of daily mean NO<sub>2</sub> concentrations, the functional relationship established with mortality is linear, thus rendering parameterisation unnecessary for their introduction into the models (Díaz et al., 1999), (Ortiz et al., 2017).

First, it is necessary to calculate the attributable risk (AR), which represents the percentage increase in daily mortality when the mean NO<sub>2</sub> concentration increases in 10 µg m<sup>-3</sup> and it is computed by using a previously estimated RR associated to this increment, via the expression:  $AR = (RR - 1)/RR * 100$  (Coste and Spira, 1991).

The RR for the municipality of Alcobendas is assumed to be the same as that estimated for Madrid, taking into account that this town is located in the northern area of the metropolitan belt of the Madrid city and, as in the case of the capital, the main source of air pollutants, including NO<sub>2</sub>, is road traffic. Relative risk is computed by Linares et al. for Madrid (Linares et al., 2018) (see Annex 1 of Supplementary Material). RR calculated for increases of 10 µg m<sup>-3</sup> in NO<sub>2</sub> levels based on natural cause mortality are found to be 1.009 (1.006; 1.013) (95% CI), in the order of the average given in the systematic review and meta-analysis published by Orellano 1.007 (1.006; 1.009) (Orellano et al., 2020). Consequently, a natural cause AR of 0.89 (95% CI: 0.58–1.28) was adopted with no significant effect found for respiratory or circulatory cause (Linares et al., 2018).

Next, the daily mortality associated with an average daily NO<sub>2</sub> concentration need to be estimated. For that, daily mean NO<sub>2</sub> concentrations are used as the measure of mean population exposure to this pollutant. Hourly available and validated data, as supplied by Ministry for the Ecological Transition and the Demographic Challenge, are taken across the period 2001–2019 at the urban traffic monitoring station of Alcobendas city belonging to the Air Quality Network of the Community of Madrid and, with comparative purposes, also at the air quality stations that constitute the Air Quality Network of Municipality of Madrid (Ministry for the Ecological Transition and the Demographic Challenge, 2022). Subsequently, taking daily mortality data (supplied by the Spanish National Statistics Institute, INE), the percentage increase in daily mortality associated with that given NO<sub>2</sub> concentration is thus calculated by multiplying it by the computed AR and dividing by the reference increase of 10 µg m<sup>-3</sup>. Afterwards, to go from the percentage increase in mortality to the number of daily deaths attributable to this NO<sub>2</sub> concentration, this percentage increase value in mortality must only be multiplied by the number of daily deaths and divided by one hundred.

Finally, it is possible to estimate the number of deaths associated with ambient concentrations of NO<sub>2</sub> registered during an investigated period by integrating the daily mortalities calculated for that required term.

## 2.3. Evaluation of attributable mortality due to NO<sub>2</sub> in ideal urban scenarios

In the first ideal scenario addressed, all roads, sidewalks and facades in the municipality of Alcobendas are considered to be covered with photocatalytic coatings and the mean NO<sub>2</sub> ambient concentration reduction is estimated. For this hypothetical scenario with reduced NO<sub>2</sub> ambient concentrations the associated attributable mortality can be computed.

In the second ideal scenario proposed, the ambient conditions correspond to an average annual NO<sub>2</sub> concentration of 20 µg m<sup>-3</sup> and in

this case the attributable mortality is also inferred. This scenario corresponds to the AQG level proposed as interim target 3 for annual mean NO<sub>2</sub> concentration by WHO (World Health Organization, 2021).

## 2.4. Cost-benefit assessment related to the implementation of photocatalytic materials in urban outdoor scenarios

Beyond the inestimable benefit of saving human lives, a very relevant aspect to consider is the economic viability of the application of photocatalytic materials, associated with both costs due to their implementation and costs saved, mainly related to the health benefits derived from the elimination of ambient NO<sub>2</sub>. In this work, a comparative estimate is presented, taking as a case study an area of 1 km<sup>2</sup> in which the street where the experiments were carried out in Alcobendas is embedded.

Recently, damage costs of main pollutants from transport have been presented by the European Commission, covering not only health effects (90% of the total external effect), but also quantifying the side effects of emitted NO<sub>x</sub> on materials (e.g. buildings), biodiversity, and crops. Existing studies on external costs have mainly concerned road transport. The evidence shows that road transport has by far the largest share in total external costs of transport. For Spain, a value of 8500 € per tonne of NO<sub>x</sub> is given (Essen et al., 2020). This would imply that each tonne of NO<sub>x</sub> withdrawn from the atmosphere would save such cost.

In this work, total NO<sub>x</sub> vehicle emissions  $E_{NO_x}$  (g h<sup>-1</sup>) in a selected 1 km<sup>2</sup> area were estimated by means of the expression:  $E_{NO_x} = EF_{NO_x} \cdot l_{roads} \cdot DMI$ , where  $EF_{NO_x}$  is the average emission factor taken as representative of the fleet traveling within that area (g km<sup>-1</sup>),  $l_{roads}$  is the total length of the main roads considered (km) and DMI is the daily mean intensity from the traffic counts registered (vehicles h<sup>-1</sup>).

Having considered that the surface deposition rates of NO and NO<sub>2</sub> are equal the estimated reduction in the ambient concentration of NO<sub>2</sub> is also applicable for NO<sub>x</sub>. Additionally, to estimate the potential benefit of reducing the ambient concentration of NO<sub>x</sub> in the estimated proportion a numerical model of atmospheric dispersion should be used (Izquierdo et al., 2020). Alternatively, the estimated ambient NO<sub>x</sub> reduction could simply be considered a consequence of reducing NO<sub>x</sub> emissions by the same proportion. The expected benefit (euros year<sup>-1</sup>) associated with the ambient NO<sub>x</sub> removed as a consequence of the application of photocatalytic materials can be calculated by applying the aforementioned external cost referred to road transport.

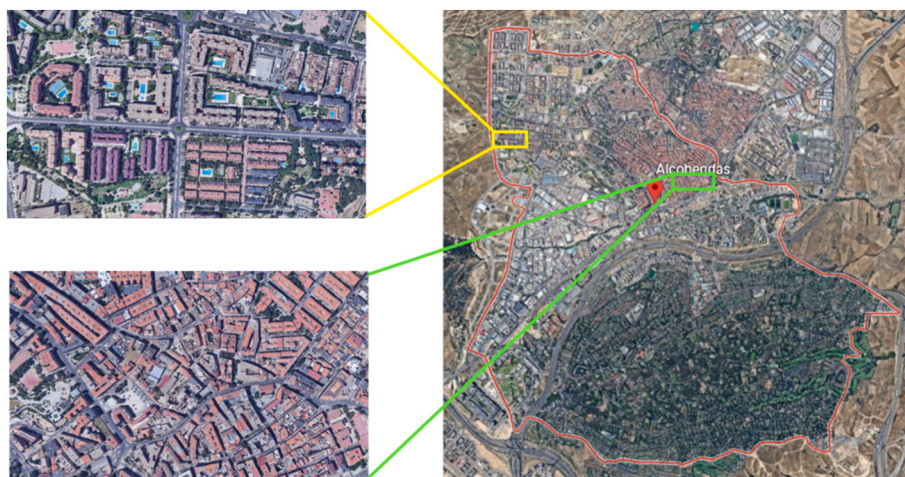
Moreover, the cost for the acquisition and setup of the products selected for their application on roads, sidewalks and facades surfaces within the area of 1 km<sup>2</sup> studied was estimated taking into account data supplied by manufacturers and public administration responsible.

## 3. Results

### 3.1. Ambient NO<sub>2</sub> removal in ideal conditions for the city of alcobendas

Upper limit photocatalytic NO<sub>2</sub>-degradation was estimated in a stretch of Paseo de la Chopera (Alcobendas) an urban main street, whose geometry nearly matches the canon of an ideal typical street canyon. The chosen street presented a cross section of 36 m wide by 16 m height. A wind direction parallel to the canyon was considered so that the air mass stream flows at a constant velocity parallel to its longitudinal axis with no back mixing. Fig. 2 shows the municipality of Alcobendas where residential areas with large avenues contrast with other narrow streets that constitute the oldest part of the city.

In the course of the development of the LIFE MINOX-STREET project, modified-ISO 22197-1:2007 laboratory assays were performed for the two different TiO<sub>2</sub>-water emulsions and the monocomponent nanopaint selected to be implemented in a road, a sidewalk and a facade of Alcobendas, respectively. NO averaged inlet concentration was set to 140 ppb instead of the standard value of 1000 ppb. The use of a classical first-order kinetic approximation to calculate the surface deposition rates of



**Fig. 2.** Map of Alcobendas. Delimitation of the municipality pointed out in red line. Experimental area of the Paseo de la Chopera (LIFE MINOX-STREET project) framed in yellow. A detail of the downtown of Alcobendas indicated in green. Images have been taken from Google Earth geobrowser. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

NO is admissible at concentrations in the range of relevant air pollution levels (Mothes et al., 2018), (Ifang et al., 2014). Thus, the estimate obtained for the selected test condition gave a result for the respective NO surface deposition velocity of  $7.2 \cdot 10^{-3}$ ,  $8 \cdot 10^{-3}$  and  $1.6 \cdot 10^{-3} \text{ m s}^{-1}$ , for the three mentioned products. Interestingly, activity was similar to those obtained for NO by other studies (Mothes et al., 2018), (Gallus et al., 2015a), (Ifang et al., 2014), (Engel et al., 2015). Nevertheless, it is important having in mind that in such ISO bed photo-reactors, transport limitations occur that can lead to underestimation of the activity by possible diffusion limitations (Ifang et al., 2014).

The average wind speed in the street canyon was assumed to be  $1.5 \text{ m s}^{-1}$ , estimated by using measurement data within the experimental campaign carried out in studied street in the course of the project LIFE MINOX-STREET (Pujadas et al., 2016), (Fernández-Pampillón et al., 2021). It was considered that air polluted mass is transported longitudinal through the canyon, without dilution to the upper atmosphere and traverses 300 m long. Under this condition, the upper limit of NO<sub>2</sub> residence time in the street could be estimated as 200 s. When the air flow is established in other directions, the turbulent mixing in the canyon increases shortening the NO<sub>2</sub> residence time in the canyon.

All the canyon surfaces (roads, sidewalks and facades) were assumed to be active. By taking estimated NO surface deposition velocities for the three mentioned selected photocatalytic materials implemented in real urban areas during the project and considering NO<sub>2</sub> uptakes to be the same as those calculated for NO, a surface reactivity given by an NO<sub>2</sub> average surface deposition velocity of  $5.6 \cdot 10^{-3} \text{ m s}^{-1}$  has been used in this study. This gives an NO<sub>2</sub> uptake coefficient of  $6.1 \cdot 10^{-5}$ . An active surface to air volume above the surface ratio ( $S_{\text{active}}/V$ ) of  $0.1 \text{ m}^{-1}$  was then taken to calculate a NO<sub>2</sub> first-order rate constant equal to  $6.6 \cdot 10^{-4} \text{ s}^{-1}$ . Considering a residence time of 200 s, the NO<sub>2</sub> degradation was estimated, leading to a maximum estimated photocatalytic NO<sub>2</sub> potential remediation of 12%, assuming all surfaces to be totally illuminated.

However, by using this approximation, no transport limitations were considered and only surface activity has been taken into account, neglecting turbulent mixing and quasi molecular-diffusion. If these latest were included, the real NO<sub>2</sub> uptake would decrease more than a factor of two (VDI, 2006). Moreover, surfaces are not active during night time period and assuming half the daytime there is enough UV-A radiation for photocatalysis to take place, a diurnal upper limit of approximately 3% is reached.

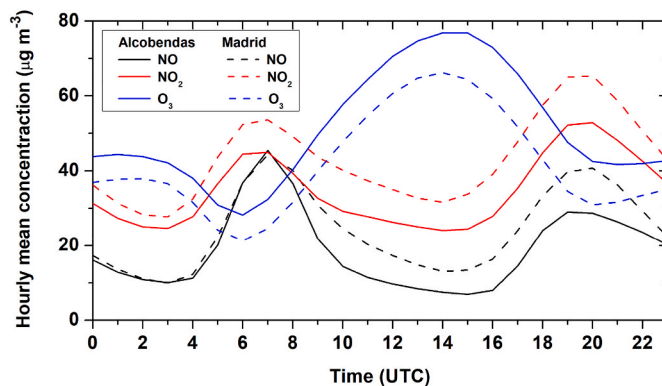
Being aware that the difference between the streets that make up the urban network of Alcobendas is enormous, that upper limit of the impact on the concentration of ambient NO<sub>2</sub> potentially generated by the use of photocatalytic materials on roads, sidewalks and facades of the whole

urban environment has been adopted intentionally to study the potential benefits of that measures on public health. For it, the estimated reduction percentage for the selected section of Paseo de la Chopera has been extrapolated to the entire municipality.

### 3.2. Attributable mortality due to NO<sub>2</sub> in a real urban scenario

Taking into account the influence of ambient concentrations of NO<sub>2</sub> on the attributable mortality to said pollutant and since the RR taken for the city of Alcobendas has been the same as that previously estimated for Madrid, the hourly average concentration of NO, NO<sub>2</sub> and O<sub>3</sub> has been calculated for the study period both in Alcobendas and in the nearby city of Madrid managing the data from the air quality stations at Alcobendas and Madrid city, for the period 2001–2019. The profiles represented in Fig. 3 clearly reflect the influence of road traffic emissions as the most relevant primary emission source in both locations, with maxima of NO and NO<sub>2</sub> corresponding to peak hours. Likewise, the photochemical generation of secondary NO<sub>2</sub> and O<sub>3</sub> is observed, with a time evolution profile in accordance with the aforementioned primary emission pattern, as reflected in previous studies related to atmospheric pollution in the Community of Madrid (Palacios et al., 2002), (Pujadas et al., 2000) (Borge et al., 2012). It should be noted that, as expected, the concentration levels of nitrogen oxides are higher in the city of Madrid, as well as its ozone levels are lower than those recorded in Alcobendas.

Moreover, the evolutions of the monthly average concentrations of NO<sub>2</sub> calculated for both Madrid and Alcobendas also show similar



**Fig. 3.** Hourly mean profile of NO, NO<sub>2</sub> and O<sub>3</sub> for Alcobendas and Madrid city concerning the period 2001–2019.

patterns, although the difference observed between the warmest and coldest months is slightly more pronounced in Alcobendas. Fig. 4 shows related statistics for both locations analysed across the study period.

Fig. 5 shows the average monthly mortality corresponding to natural cause in Alcobendas computed for 2001–2019, with an annual mean value of  $486 \pm 62$  deaths. Furthermore, the attributable mortality due to NO<sub>2</sub>, according to the RR assumed for Alcobendas, is presented in the lower panel.

The highest values were observed in the fall and winter seasons. Although the annual evolution of total mortality reflects a slight seasonal influence, meteorology is clearly a determining factor on the recorded levels of NO<sub>2</sub> (Fig. 4). Indeed, during these colder months, the Community of Madrid is frequently under the synoptic influence of high-pressure systems, associated with poor ventilation events and strong surface thermal inversions. All these factors, together with the limited photolysis of NO<sub>2</sub>, lead to the accumulation of this pollutant in ambient air and the development of local episodes of urban pollution (Pujadas et al., 2000). This circumstance is reflected in the higher NO<sub>2</sub> attributable mortality found for Alcobendas during the colder seasons. Integrated attributable mortality due to NO<sub>2</sub> concentrations for Alcobendas for the study period was 289 deaths, representing a 3.1% of total attributable mortality due to natural cause for this municipality.

Fig. 6 shows the annual evolution of the two factors that determine the attributable mortality due to NO<sub>2</sub> for the chosen period. Until 2008, the annual average concentration of NO<sub>2</sub> increased, with a mean value around  $40 \mu\text{g m}^{-3}$ , but from that year on it stabilized at lower values, around  $30 \mu\text{g m}^{-3}$ , and even decreased slightly. On the other hand, total mortality due to natural cause normalized to 1000 inhabitants shows a growing trend throughout the period studied, more pronounced as of 2008, which cannot be attributed exclusively to population growth, but probably also to its progressive aging (Fig. 7) and to other influencing factors (Murray et al., 2020).

Clearly, during the period discussed, there has been a progressive increase in the mortality due to natural causes attributed to age groups over 80 years old, which doubled their population in 2019 compared to the value recorded in 2001. Thus, population distribution must therefore be understood as a key factor, determinant of mortality from natural causes and, therefore, of mortality attributable to NO<sub>2</sub> in a specific location.

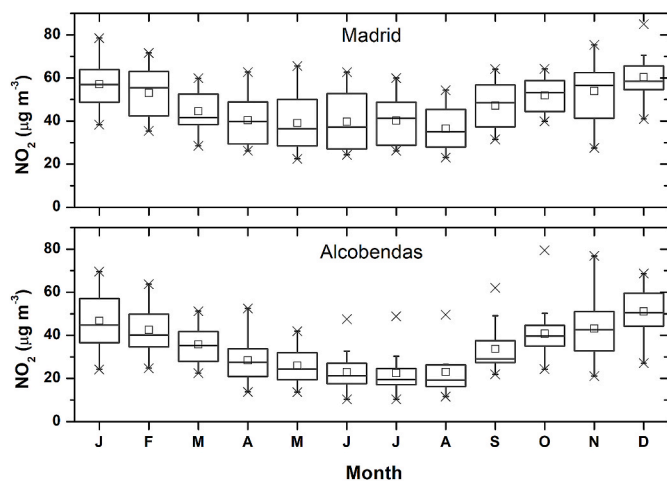


Fig. 4. Box plots for NO<sub>2</sub> concentrations registered by the air quality station at Alcobendas (Air Quality Network of Community of Madrid) and air quality stations at Madrid city (Air Quality Network of Municipality of Madrid) during the period 2001–2019. Box: lower and upper limits are the 25th percentile (Q1) and the 75th percentile (Q3), respectively; median (line); mean (open symbol). Upper/lower bars: the largest/lower observed point from the dataset that falls within the distance of 1.5 times the interquartile range. Cross: maximum and minimum.

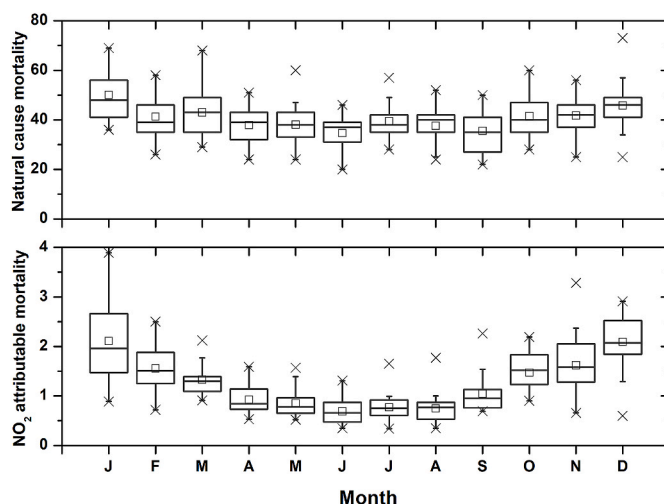


Fig. 5. Upper: Monthly mortality due to natural cause. Lower: Monthly attributable mortality due to NO<sub>2</sub>. Box: lower and upper limits are the 25th percentile (Q1) and the 75th percentile (Q3), respectively; median (line); mean (open symbol). Upper/lower bars: the largest/lower observed point from the dataset that falls within the distance of 1.5 times the interquartile range. Cross: maximum and minimum. Figures refer to Alcobendas municipality during the period 2001–2019.

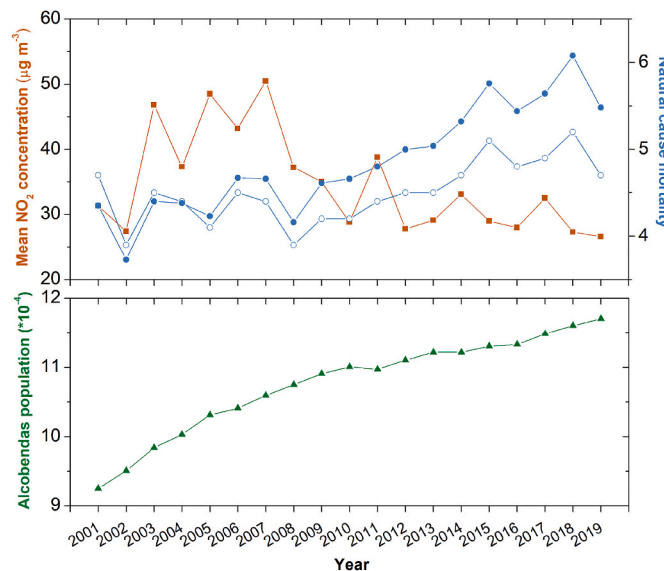


Fig. 6. Upper: Annual mean NO<sub>2</sub> concentration (left axis, square); total mortality due to natural cause  $\times 10^{-2}$  (right axis, solid circle); total mortality due to natural cause per 1000 inhabitants (right axis, open circle). Lower: Alcobendas population (triangle). Data furnished by INE. Figures refer to Alcobendas municipality during the period 2001–2019.

### 3.3. Attributable mortality due to NO<sub>2</sub> in an ideal urban scenario

Considering a 3% reduction in ambient mean NO<sub>2</sub> concentrations due to the hypothetical massive implementation of selected photoactive products on the roadways, sidewalks and facades of Alcobendas city, attributable mortality due to NO<sub>2</sub> concentrations would be 280 deaths. Furthermore, provided that ambient NO<sub>2</sub> values met the  $20 \mu\text{g m}^{-3}$  threshold, a 57% reduction in total number of deaths with respect to that estimated for reference scenario was found (see supplementary material, Table 1). Fig. 8 shows the attributable mortality due to NO<sub>2</sub> concentrations for those latest two cases in which NO<sub>2</sub> concentrations are taken to be reduced with respect to the reference scenario.

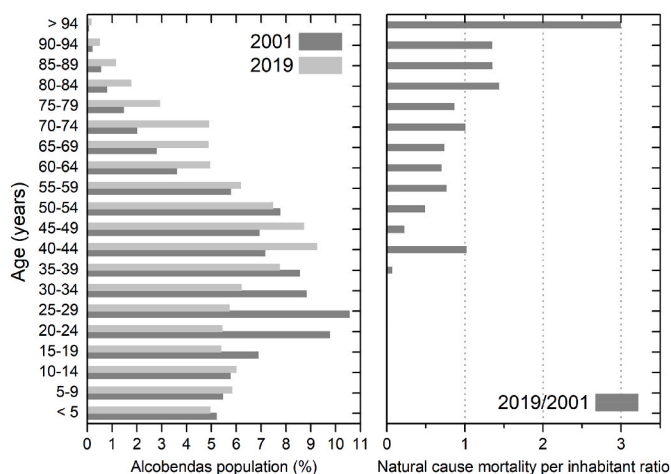


Fig. 7. Age distribution of population in 2001 and 2019 (%) (left) and natural cause mortality per inhabitant ratio 2019 vs. 2001 (right), in Alcobendas.

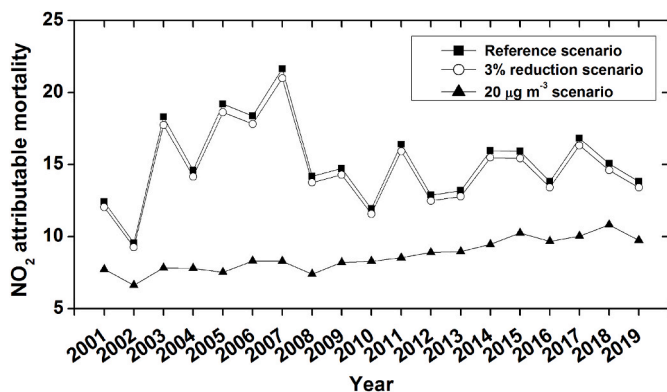


Fig. 8. Attributable mortality due to NO<sub>2</sub> concentrations for Alcobendas during 2001–2019 for the investigated ambient NO<sub>2</sub> reduction scenarios (3% and 20 µg m<sup>-3</sup> threshold) compared to the reference one.

### 3.4. Cost-benefit estimation for the 3% NO<sub>x</sub> reduction scenario

In the course of a NO<sub>x</sub> measurement campaign carried out in Alcobendas, traffic volume in the selected stretch of Paseo de la Chopera street was also exhaustively determined on three different days throughout the measurement period (September 29th, October 15th and 22nd, 06:00–17:00 UTC) by using a video camera. An average of 610 ± 113 vehicles h<sup>-1</sup> was registered at the Paseo de la Chopera. Traffic counts were analysed by category of vehicles (for which passenger cars represents 85.3% of the fleet, with 9% vans, 2% buses, 1.6% trucks and 2.1% motorcycles). An average speed of less than 60 km h<sup>-1</sup> was found for the 90% of the fleet (data was supplied by the Police of the municipality of Alcobendas) (Fernández-Pampillón et al., 2021). In order to estimate the total NO<sub>x</sub> emissions within an area of 1 km<sup>2</sup> around the selected street, an average emission factor for NO<sub>x</sub> of 0.544 g vehicle<sup>-1</sup> km<sup>-1</sup> (NO<sub>x</sub> expressed as NO<sub>2</sub>) was used (Ait-Helal et al., 2015). A total length of approximately 8 km was considered taking into account all the main roads enclosed in that area. Finally, a total NO<sub>x</sub> emission of 7.1 10<sup>-7</sup> tonne s<sup>-1</sup> has been estimated.

The reduction of 3% in the ambient NO<sub>x</sub> concentration gives a total of 0.67 tonne NO<sub>x</sub> yr<sup>-1</sup> withdrawn from the atmosphere as a result of the application of the photocatalytic materials. By applying the mentioned damage cost of NO<sub>x</sub> from transport, a benefit of 5708 euros yr<sup>-1</sup> could be derived.

Additionally, the cost for the acquisition and setup of the products selected for their application on roads, sidewalks and facades surfaces

within the area of 1 km<sup>2</sup> studied (50000 m<sup>2</sup>, 550000 m<sup>2</sup> and 400000 m<sup>2</sup>, respectively) (data supplied by municipality of Alcobendas) was estimated. In Table 1, cost of the photocatalytic materials purchase and its application is given. Therefore, a total cost of k€4750.5 can be derived.

Obviously, cleaning should be taken into account in the maintenance costs. However, selected materials do not require additional or different cleaning practices from the usual ones carried out in the municipality, reason why these costs have not been considered in this evaluation.

## 4. Discussion

The existence of an association between ambient NO<sub>2</sub> and cardiovascular and all-cause mortality has been demonstrated for the population of some Spanish cities (Saez et al., 2002). More recently, Linares et al. (Linares et al., 2018) have updated the impact of daily mean NO<sub>2</sub> concentrations on population mortality at a national level analysing the short-term association between NO<sub>2</sub> concentrations and natural-cause, circulatory-cause and respiratory-cause mortality in all Spanish provinces across the period 2000–2009.

Although the technology implemented in European cities in order to reduce emissions, specially from road traffic, has allowed levels of air pollution to have been progressively decreasing in recent decades (Querol et al., 2014), (Colette and Rouil, 2020), pollutant sudden increases from their background levels, known as high pollution episodes, continue to occur. These episodes are highly conditioned not only by the intensity of the emissions but also by the highly stable atmospheric conditions produced under specific synoptic meteorological patterns associated with high pressure systems (Artiñano et al., 2003), (Kukkonen et al., 2005), (Reizer and Juda-Rezler, 2016), (Largerón and Staquet, 2016), (Borge et al., 2018). Particularly in the Community of Madrid, these systems, frequently located near the Iberian Peninsula, generate strong atmospheric stability in winter and autumn. And, as a consequence, the development of frequent thermal inversions, with light winds and low turbulent vertical exchange. These conditions give rise to low-dispersion local circulations (Pujadas et al., 2000), (Martilli et al., 2021), (Salvador et al., 2021), (Valverde et al., 2015), that produce episodes, sometimes of several consecutive days, of high NO<sub>2</sub> pollution in urban areas, above of the limit levels established for this pollutant. An analysis of the situation of Alcobendas, taking into account the guideline values for NO<sub>2</sub> levels, across the studied period 2001–2019, has revealed a worrying situation in which mean annual concentrations exceeded the EU annual limit in the 21% of the analysed years. During the months of September to March, the average monthly concentration of NO<sub>2</sub> observed was a factor 1.7 higher than that registered during the period from April to August, due to the different climatic-meteorological (dispersive) and emission conditions (see supplementary material, Table 2).

Given that the susceptibility of the populations to adverse health effects of air pollution may be quite different from one place to another, an adequate choice of dose-response function and appropriate determination of population exposure levels are crucial to quantify the impact of any atmospheric pollutant on health. Spanish values for relative risks calculated for increases of 10 µg m<sup>-3</sup> in NO<sub>2</sub> levels based on natural cause mortality are in the range from 1.007 (95% CI: 1.005–1.009) to 1.051 (95% CI: 1.016–1.086), showing a wide variability (Linares et al., 2018). In the present study, the relative risk taken for Madrid has been supposed to represent the dose-response function needed to estimate the

Table 1

Materials purchase and application costs of the photocatalytic products selected for their implementation in urban surfaces of the area selected in Alcobendas. Data supplied by the Municipality of Alcobendas (2018).

Cost (€ m <sup>-2</sup> )	Road	Sidewalk	Facade
Material	1.25	1.08	2.38
Application	1.00	4.00	2.23

attributable to NO<sub>2</sub> mortality for Alcobendas, considering that no relevant differences in pollutant mix, concentration levels, disease composition and prevalence levels, and susceptibility of underlying population exist between these two cities.

On the other hand, the seasonal variation factor of total mortality assessed for the coldest versus warmest periods was 1.1 (see supplementary material, Table 3). This fact, together with the variability just mentioned in ambient NO<sub>2</sub> concentrations, is reflected in the annual evolution estimated of the mortality attributable to NO<sub>2</sub> that presents a seasonal variation factor of 2 (see supplementary material, Table 4).

Although a priori the choice of a RR for Alcobendas identical to that for Madrid is reasonable, there are factors that could affect the relationships between observed air pollution and daily mortality (Barceló et al., 2016) that could not be controlled by Linares et al. (Linares et al., 2018). These authors distinguished the effects of NO<sub>2</sub> per se from the modifying effects of PM<sub>10</sub>, but daily levels of other pollutants such as SO<sub>2</sub> or O<sub>3</sub> were not investigated as possible confounding. Moreover, the effect of certain explanatory variables at the individual level, such as sex, age and residence of the subjects or other confounders such as individual socioeconomic data, lifestyle, and comorbidities, which may differ among people residing in different locations were not considered (Vodanos et al., 2015), (Samoli et al., 2006).

Even when the average daily concentrations of NO<sub>2</sub> were similar for different locations, those mentioned factors will decisively influence daily mortality, so that, for a selected RR, the mortality attributable to NO<sub>2</sub> may differ significantly. In fact, while the annual mean mortality attributable to NO<sub>2</sub> concentrations for Alcobendas during the study period was 1.4 deaths/10000 inhabitants, that corresponding to the city of Madrid was 3.4 deaths/10000 inhabitants. This is due both to the ratio found in the average annual concentration of NO<sub>2</sub> between Madrid and Alcobendas (1.3), and to the fact that the average annual mortality per 10000 inhabitants in Madrid was approximately twice higher than the figure found for Alcobendas (see supplementary material, Table 5).

Concerning the reported efficiency of NO<sub>x</sub> reduction on TiO<sub>2</sub>-treated surfaces, several field experimental studies have been developed at real scale in outdoor conditions whose results have yielded quite low to non-detectable reductions (IPL, 2010), (TERA environment, 2009), (Gallus et al., 2015a), (Gallus et al., 2015b), (Barratt et al., 2012). Other studies done under unrealistically high S<sub>active</sub>/V ratios have shown estimated reductions of only ~5% when scaling down to real urban street conditions (Laufs et al., 2010), (Maggos et al., 2008), (Moussiopoulos et al., 2008), (Fraunhofer Institute for Molecular Biology and Applied Ecology, 2009). Similar results (<3%) were found in an urban area of Alcobendas in which a selected photocatalytic TiO<sub>2</sub>-based water emulsion was applied on a bituminous pavement (Fernández-Pampillón et al., 2021) in the framework of LIFE MINOX-STREET project. The latest value is in the order of the estimate presented here for the municipality of Alcobendas.

In this study, NO<sub>2</sub> surface deposition velocities utilized to compute the NO<sub>2</sub> potential degradation in Alcobendas have been taken to be equal to NO uptakes estimated for the three selected photoactive materials implemented in this city by using standard 22197-1:2007 test, that applies only for NO remediation. As long as similar uptakes for both NO<sub>x</sub> pollutants or even up to one order of magnitude lower for NO<sub>2</sub> have been reported for most studies, this assumption was considered to be valid in order to estimate a maximum NO<sub>2</sub> surface deposition flux. However, the selected NO<sub>2</sub> deposition rate could be obviously over-estimated such that if the NO<sub>2</sub> uptake chosen had been an order of magnitude lower, the environmental NO<sub>2</sub> reduction and, consequently the benefits in terms of reduction in mortality due to NO<sub>2</sub> would have decrease, approximately, in the same extent. The evaluation of the NO<sub>2</sub> efficiency in a more realistic way continues to be a research area focused in designing specific laboratory tests that pay special attention to transport phenomena limitations and the NO<sub>2</sub> gas phase relevant photochemistry (Ifang et al., 2014).

Assessing the impact of this technology in the urban air quality has

been tackled here by using a first-order kinetic approach that has allowed the estimation of maximum NO<sub>2</sub> expected removal efficiency under particular atmospheric conditions at an urban area. This simplification must be used only when the chosen street behaves as a canyon under specific ambient conditions so that constant air mass flow goes parallel to the longitudinal axis and well-mixed atmospheric situation avoids concentration gradients. Nevertheless, taking into account other atmospheric crucial phenomena, as turbulence or transport limitations, microscale models could be used to better define pollutants dispersion and concentration fields.

It must be emphasized that the estimation of the ambient NO<sub>2</sub> removal capacity of the photocatalytic technology applied to the city of Alcobendas is an ideal upper limit since it has been carried out under optimal conditions (unrealistic) and other factors, which decisively influence the photocatalytic activity, have not been taken into account (orientation of the streets that prevents all urban surfaces from being illuminated simultaneously; width/height relationships of the streets that determine the shadows casted by buildings and obstacles; variation in environmental conditions such as concentration and emission of pollutants, solar irradiance, speed and direction of wind or relative humidity; wetted conditions; aging, soiling or wearing of active areas).

In Alcobendas, the streets of the downtown area, that represent a 7.5% of the whole urban grid (Google Earth Pro tools), have characteristics that would decrease daily photocatalytic activity. They are narrow, which dramatically reduces the potential of the surfaces under consideration to be illuminated. Furthermore, their average S<sub>active</sub>/V ratio is also low, limiting the heterogeneous NO<sub>2</sub> uptake. In addition, the traffic flow can be increased by the frequent congestions that occur, reducing the potentially active area. Finally, its irregular layout would favour turbulent mixing, reducing the residence time of street air masses and, consequently, their possibility of interaction with photocatalytic surfaces.

Taking into account the low NO<sub>2</sub> removal potential due to the use of TiO<sub>2</sub>-based photocatalytic materials in outdoor urban scenarios and that the estimated damage costs potentially saved by the use of this technology could be negligible compared to the expected acquisition and installation costs, this type of strategy alone cannot solve air quality problems. In addition to advance in the research of new photocatalytic materials and new engineering designs, it is key that in those future applications the photocatalytic materials can be used under controlled conditions and with a significant increasing of the S<sub>active</sub>/V ratio (Huang et al., 2021). Their efficiency should be further investigated in order to determine to what extent their combined use with other abatement measures, targeted to the road traffic sector source and already considered in the urban air quality plans (e. g. definition of low emissions zones, reduction of road capacity and pedestrianized areas in the city centres, renovation of city bus fleets to incorporate cleaner technologies, promotion of the use of cleaner vehicles and the public transport, etcetera) (Ministry of Agriculture and Fishing and Food and Environment, 2017), (Borge et al., 2018), (Municipality of Madrid, 2019), could contribute achieving the fulfilment of NO<sub>2</sub> air quality standards and noticeably reduce the attributable mortality to NO<sub>2</sub> in urban areas.

## 5. Conclusions

Within the framework of the European LIFE MINOX-STREET project, the potential usefulness of a variety of commercial photocatalytic materials to act as ambient NO<sub>x</sub> sinks when implemented on urban surfaces was evaluated. A wide variety of laboratory-scale experiments were developed in order to select three of the tested materials for their implementation in real urban scenarios in the city of Alcobendas (Community of Madrid). In this work, the results obtained from testing the photocatalytic activity of the selected materials, in outdoor conditions by using a modified standard method, were subsequently utilized to derive the corresponding surface NO<sub>x</sub> deposition velocities following a first-order kinetic approximation.



Realistic NO<sub>x</sub> surface deposition velocities are essential in order to assess which could be the ambient effect on pollution levels if photocatalytic materials were applied in a particular area or, furthermore, to model the foreseen effects if such materials were implemented in a whole city. Particularly, when upper limit photocatalytic NO<sub>x</sub>-degradation was estimated in an urban main street canyon of Alcobendas, based on the NO<sub>x</sub> deposition velocities obtained in the present study under optimal ambient conditions, a reduction of only a few per cent was found (3%).

The application to the municipality of Alcobendas of the dose-response function relating NO<sub>2</sub> ambient concentrations and associated mortality for Madrid city, has allowed estimating the mortality attributable to that pollutant in that municipality over an extended period. A marked seasonal dependence has been observed, function of both the climatic-meteorological and air pollutant emission conditions with a minimum in NO<sub>2</sub>-attributable mortality in the spring-summer period.

For a more precise study of the benefit that the implementation of the mentioned technology would have in Alcobendas in terms of deaths avoided, it would be necessary to have an individualized estimate of the RR for the municipality and, in addition to the explanatory variables considered in the proposed methodology, it would be necessary to consider other factors as relevant as the characteristics of the distribution of the population and socioeconomic conditions.

Finally, the cost-benefit evaluation associated with the application of photocatalytic materials in Alcobendas yields results that question the profitability of using this technology as a NO<sub>2</sub> depolluting strategy, with the relative cost of acquisition and implementation notably higher than the savings derived from the maximum potential decrease in the calculated atmospheric NO<sub>2</sub> concentration. Additionally, and although it falls outside the scope of this work, it would be highly recommended to carry out a comparative analysis, in terms of both efficiency and cost-benefit balance, of this strategy with other NO<sub>2</sub> remediation options included in urban air quality plans. And, subsequently, to analyse their complementarity and to efficiently help air quality managers to protect the population, specially in particularly sensitive areas.

#### Credit author statement

Jaime Fernández-Pampillón: Conceptualization, Methodology, Formal analysis, Investigation, Data curation, Writing – original draft, Writing – review & editing, Project administration, Funding acquisition. Magdalena Palacios: Conceptualization, Methodology, Formal analysis, Investigation, Data curation, Writing – original draft, Writing – review & editing, Project administration, Funding acquisition. Lourdes Núñez: Conceptualization, Methodology, Formal analysis, Investigation, Data curation, Writing – original draft, Writing – review & editing, Visualization. Manuel Pujadas: Conceptualization, Methodology, Formal analysis, Investigation, Writing – original draft, Writing – review & editing, Supervision. Begoña Artñano: Conceptualization, 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.

#### Data availability

Data will be made available on request.

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#### Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.envpol.2023.121203>.

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