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REVIEW ARTICLE

Removing NO_x Pollution by Photocatalytic Building Materials in Real-Life: Evaluation of Existing Field Studies

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ARTICLE HISTORY

Received: August 13, 2020
Revised: February 03, 2021
Accepted: February 04, 2021

DOI:
10.2174/2665976X02666210308151731

Abstract: The NO_x gasses (NO and NO_2) are among the most important air pollutants due to the toxicity of NO_2, as well as the role of NO_x in the tropospheric oxidation of Volatile Organic Carbons (VOCs), contributing to the formation of other hazardous air pollutants. Air pollution is one of the biggest health threats worldwide; hence reducing NO_x levels is an important objective of the UN sustainable development goals, *e.g*., #3, “Good health and well-being” and #11 “Sustainable cities and communities.”Photocatalysis using $TiO_2$and light is a promising technique for removing NO_x along with other pollutants, as demonstrated on a laboratory scale. Furthermore, a long-range of real-life test studies of varying scales have been conducted during the past two decades. The results of these studies have been conflicting, with some studies reporting no effect on the ambient air quality and others reporting significant reductions of NO_x levels. However, the studies are very difficult to compare and assess due to the different approaches used, which consequently vary in quality. In this review, we aim to develop a set of objective evaluation criteria to assess the quality of the individual studies in order to simplify the interpretation and comparison of the existing studies. Moreover, we propose some guidelines for future test studies. Furthermore, the approaches and main conclusion from 23 studies are independently assessed and discussed herein.

Keywords: Photocatalytic building materials, titanium dioxide, nitrogen oxides, NO_x removal, field studies, evaluation criteria.

1. INTRODUCTION

According to the World Health Organization (WHO), 9 out of 10 people breathe air containing high levels of pollutants, while outdoor air pollution annually causes 4.2 million premature deaths [1]. Adverse health effects related to long-term exposure to polluted air include respiratory issues, cardiovascular diseases, and lung cancer, among others [2-4]. Moreover, air pollution causes severe damage to vegetation as well as buildings [5, 6]. In 2015, 193 countries adopted the Sustainable Development Goals (SDG) aiming to build a better world for people and our planet by 2030 [7]. Air pollution is specifically mentioned as a central concern in SDG 3.9 and SDG 11.6 and is furthermore closely interlinked with several of the remaining SDGs [8]. Among the most common air pollutants, besides Volatile Organic Compounds (VOCs), sulfur oxides (SO_x), particulate matter (PM_{10} and $PM_{2.5}$), and ozone, are the NO_x gasses, NO and NO_2 [9]. Sources of tropospheric NO_xalso include natural sources; however, anthropogenic combustion of fossil fuel is by far the most important source, with car engines accounting for 40 % of anthropogenic emissions in 2005 [10]. Long-term exposure to high levels of NO_2 has been linked to mortality, lung cancer, and respiratory problems in asthmatic children [3, 11, 12], but it is still being discussed whether these effects are specifically caused by NO_2 or by coexisting pollutants [12, 13]. In the troposphere, NO_x chemistry is closely related to the formation of ozone, photochemical smog, and secondary organic aerosol (SOA) due to its role in the oxidation of VOCs [14]. The only significant sink of tropospheric NO_x is the reaction of NO_2 with OH-radicals to form nitric acid [14], which is one of the main contributors to acid rain [15]. The EU Directive specifies that the annual mean concentration of NO_2 must not exceed 40 mg/m^3, while an hourly limit of 200 mg/m^3 must not be exceeded to more than 18 occasions each year [16]. However, the limits are exceeded on a regular basis in many locations [17, 18]. Limiting the combustion of fossil fuel is the main objective in order to ensure pure air in the future. However, transitioning fully to renewable energy has proven to be an immense political challenge of global proportions and is unlikely to happen in the near future [19]. Meanwhile, air purification technologies are essential tools to limit the consequences of poor air quality on human health and well-being as well as the societal economy [4, 20]. In this prospect, the development of NO_x removal strategies has been the main focus [21]. Photocatalysis by titanium dioxide, TiO_2, which was first discovered by Fujishima and Honda [22-24], is a promising method for removal of

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NO_x, as well as other pollutants, such as SO_2, VOCs [25-29] and even for reduction of CO_2 into solar fuels [30]. In this review, we focus on the use of TiO_2 as a photocatalyst in NO_x removing construction materials [31-34], which in a recent report was highlighted as one of the cheapest options for NO_x removal [35]. Laboratory studies have demonstrated the NO_x removing properties of TiO_2 containing building materials, such as concrete, asphalt, and paints [36-39]; however, large-scale field studies are needed in order to assess the effect on the air quality under real conditions. Several such studies have been conducted in this regard with somewhat contradicting results. In this review we aim to objectively evaluate the quality of the existing field studies. In this regard we attempt to establish some objective evaluation criteria to readily asses the level of information that can be extracted from the studies.

1.1. Photocatalytic Principle

The photocatalytic properties of TiO_2 are based on the semiconducting properties of the crystalline form of the mineral. TiO_2 exists in three polymorphs, anatase, rutile, and brookite; however, the anatase form is the most widely used for photocatalytic purposes [40]. The pure TiO_2 can furthermore be chemically modified to enhance the catalytic activity. The many modification approaches and the properties of various different TiO_2 based materials are discussed in detail in a review by Tsang *et al.* [41]. The complex process of NO_x absorption and degradation at the TiO_2 surface is described in detail elsewhere [42, 43]. Here we briefly outline the simplified mechanism. When the TiO_2 catalyst is illuminated by light with energy higher than the bandgap (3.2 eV for anatase, corresponding to UV light), transfer of an electron from the valence band to the conduction band is induced, resulting in the formation of an electron-hole pair. The resulting electron can react with adsorbed oxygen from the air to produce O_2^-. In the presence of water, the high reactivity of the hole, h^+, enables electron transfer from OH^- ions, resulting from hydrolysis of adsorbed water.The product of this reaction is hydroxyl radicals, OH:

$$OH^- + h^+ \rightarrow OH^\cdot \quad (1)$$

In the following oxidation process at the catalyst surface, which is similar to the tropospheric NO_x removal reaction, NO_x is converted to nitrate, NO_3^-, in three steps [43]:

$$NO + OH^\cdot \rightarrow HNO_2 \quad (2)$$

$$HNO_2 + OH^\cdot \rightarrow NO_2 + H_2O \quad (3)$$

$$NO_2 + OH^\cdot \rightarrow NO_3^- + H^+ \quad (4)$$

The resulting nitrate is extremely soluble in water and can, in principle, be washed away. In the case of cementitious materials, some of the resulting nitrates remain on the surface and may be a cause of reduction of the photocatalytic activity. Since the main catalytic property of the TiO_2 surface is the effective formation of OH radicals, the process is not limited to NO_x removal but also catalyzes the removal of other pollutants such as VOCs and SO_x.

1.2. NO_x Detection by Chemiluminescence

Many techniques for monitoring ambient NO_x levels have been developed, among which the chemiluminescence technique (CLD) is the most commonly used [44]. CLD NO_x monitors typically alternate between two modes: one that measures the concentration of NO by sampling ambient air directly. NO is the reacted with O_3as in Reaction 4 [44, 45]:

$$NO + O_3 \rightarrow NO_2^* + O_2 \quad (5)$$

When the resulting excited NO_2 molecule (NO_2^*) is deactivated, light in the infrared spectrum is emitted, with an intensity proportional to the NO concentration. In the second mode, the sum of NO and NO_2(NO_x) is measured. Ambient air is passed over a catalyst (usually gold or molybdenum oxide, often heated) to convert NO_2 to NO, which is subsequently oxidized by O_3 (Reaction 4). The difference between the two values is reported as the NO_2 concentration. Whereas the CLD method quantifies NO concentrations with high accuracy, theNO_x, and thereby NO_2, concentrations measured by this technique are highly uncertain [46]. It has been shown that interference of other nitrogen compounds in the air, mainly HNO_3, which are also oxidized in Reaction 4, may lead to severely overestimated NO_2 concentrations. It was found in a study by Dunlea *et al.* that the NO_2 concentration was overestimated by 50 % [44]. Alternative methods used to detect NOx in urban space include passive samplers, such as but not limited, to Passam and Ogawa tubes [47] and new low-cost air quality sensors. However, for both passive samplers and low-cost air quality sensors, careful calibration against a more accurate method is required, *e.g.*, the chemiluminescence technique, as the variance in these methods is sometimes equal to the effect obtained from photocatalysis [48]. Furthermore, when using passive samplers, the data frequency is significantly lower than when using samplers with time resolution down to seconds.

2. EVALUATION SCHEME

Reported trial studies are evaluated based on a scheme suggested herein. Hence, we define nine criteria as described below, and the studies are evaluated against these parameters. While each fulfilled parameter gives a grading of one star, nine stars in total can be obtained by using this model. We acknowledge that some of the parameters are somewhat subjective (*e.g.*, defining a minimum test site area). Hence, the evaluation is not a conclusive ranking of the studies, and it should rather be viewed as an easy overview to help to assess the level of information that can be extracted from each study.

2.1. Criterion 1: Laboratory Tests

The very basic of a successful trial study is to use a catalytic product of sufficient activity. Hence, the activity of the product should initially be tested and reported to ensure that the product is actually functioning (*e.g.*, on a laboratory scale). The most widely recognized test procedure applied to determine the activity of NO_x removing building materials using photocatalysis is the ISO 22197-1 standard test [49]. This standard utilizes the flow-through method, in which 1 ppmV NO gas passes the sample while illuminated by a UV lamp with an intensity of 10 W/m^2 in the wavelength range of 300-400 nm. The NO_x concentration is measured at the inlet and outlet.To allow comparability of different studies, tests should be conducted using the same procedure, preferentially a standard test such as the ISO 22197-1 method, which is the most widely used. It is, however, noted that criticism of the ISO method had been reported in the literature [50, 51]. Therefore, the studies evaluated here will obtain a star for any reliable, initial laboratory test. Other test methods include the UNI and CEN methods [52, 53]. Note that photocatalytic activities obtained under different laboratory conditions are not directly comparable, as the activity is highly dependent on factors such as light intensity, relative humidity, *etc*. [37, 54].

2.2. Criterion 2: Test Site Area

The area of the photocatalytic site should be large enough to have a measurable impact on the ambient NO_x concentration. Defining a specific minimum test site area presents a challenge. While in some studies, performed under very controlled conditions, a smaller area may be sufficient, in most cases, the larger the area, the more reliable results can be expected. As there are, to the best of our knowledge, no studies investigating the effect of the test site area, we assign a star if the total area is larger than 250 m^2. This limit is chosen so that no studies of very similar areas are excluded. We note that this area, in many cases, is still relatively small, and even larger test areas are likely to give more reliable results. Furthermore, the test site geometry is an important parameter, as a larger active area to volume ratio is expected to result in a larger effect on air quality.

2.3. Criterion 3: Monitoring Distance

The measurable effect of a photocatalytic surface will significantly depend on the distance from the surface at which the NO_x level is detected. Hence, it is crucial to report this distance. Typically, this is the monitoring height, as most studies consider a ground-level surface, such as a pavement or a road. Here, we recommend that air sampling should be performed at a monitoring height that realistically reflects human exposure to assess the effect on the ambient air quality. Hence, a star is assigned for monitoring distances between 0.5 m and 3 m.

2.4. Criterion 4: Reference Data

Monitoring data from the trial site should be compared to simultaneous monitoring data from a reference site closely resembling the test site. It is crucial to compare the test site data with reference data, also when a blank test is performed, in order to take into account potential changes over time in NO_x emissions, *etc*. The blank test should clarify if the initial NO_x levels are similar at the reference site and the test site. The reference data can be supplemented by day/night measurements or light on/light off data in the case of tunnel studies; however, also these data should, ideally, likewise be performed at the test site and the reference site simultaneously.

2.5. Criterion 5: Blank Data

Initial monitoring before construction of the photocatalytically active material should be conducted to obtain reliable blank data. This should be performed at the reference site and the test site to clarify potential differences between the two sites. The monitoring should ideally take place in a similar period of time as the test campaign, and the duration of the blank test should be long enough to ensure statistically reliable blank data. Blank data should also be collected when a reference site is used to minimize uncertainties relating to the difference between NO_x levels, *etc*., at the two sites.

2.6. Criterion 6: Duration of Study

The study should have a duration that is long enough to take into account fluctuations in NO_x levels, meteorological parameters, *etc*. Ideally, the duration should be several months. Here, a star is obtained if the duration of the active period is more than a month. Note that comparison of studies performed in, *e.g*., winter and summer may not be comparable. Likewise, differences in local climate may complicate comparison between studies.

2.7. Criterion 7: Monitoring Frequency

Sampling should be performed with sufficient frequency to ensure enough data points for statistical evaluation of data. Ideally, continuous monitoring should be performed, alternatively, and we consider that measurements should be carried out more frequently than once a day throughout the project. In this case, it is crucial that the blank measurements and the test measurements are collected at the exact same time of the day, while the reference data and test site data should be measured simultaneously or at least within a very short time gap, preferentially within minutes, due to concentration variations during the day.

2.8. Criterion 8: Durability Testing

While the initial activity test of the material ensures that the product has the potential to remove NO_x, a high initial activity does not guarantee persistent activity, in particular for studies of a longer duration. Thus, the photocatalytically active material should, during the trial study, periodically be subjected to activity testing in the laboratory to ensure persisting activity of the product. Alternatively, the photocatalytic material can be subjected to durability testing in the laboratory.

2.9. Criterion 9: Supplementary Data

In addition to the NO_x levels, the studies would benefit from supplementary data, including meteorological data, traffic counts, and other relevant data. In a study by Dylla and Hassan, it was shown that even small changes in vehicle activity at the active site and the reference site could result in significant differences in NO_x emission, complicating interpretation of NO_x abatement results [55]. Interpreting many parameters which potentially influencing the measurements generally presents a great challenge. Promising attempts to develop an artificial intelligence model to evaluate the field performance, taking into account a long range of parameters, have been made [56]. Monitoring supplementary data is of immense importance, in particular where a reference site is not available.

3. DISCUSSION

In this review, 23published field studies have been evaluated and discussed according to the criteria given in Section 2. The results are shown in Table **1**. In some cases, information about the test site design has been left out of the

Table 1. Overview of the evaluation of 23 field studies according to the criteria explained in Section 2.

Study[a]	Lab	Area	Distance	Ref.	Blank	Duration	Frequency	Durability	Suppl.
Antwerp	★	★	-	★	-	★	-	★	★
Guerville	-	-	★	★	-	★	★	-	★
London	-	-	★	-	★	★	★	-	★
Rome	★	★	★	★	★	-	★	-	★
Hengelo	★	★	★	★	★	★	-	★	★
Malmø	-	★	★	★	★	★	★	-	★
Manila	-	★	-	-	★	★	-	-	★
Brussels	★	★	-	★	★	★	★	★	★
Louisiana	★	-	-	-	★	-	★	★	★
Wijnegem	★	-	-	-	-	★	-	★	-
Gasværksvej	★	★	★	★	★	★	★	-	★
CPH airport	★	★	★	★	★	★	★	-	-
The Hague	★	★	-	-	-	-	★	-	★
Fælledvej	-	★	★	-	★	★	★	-	★
Holbæk mv	-	-	-	★	-	★	★	-	★
Valencia	-	★	★	-	-	-	★	-	-
Toronto	-	-	-	-	-	★	★	-	-
Roskilde	★	★	-	-	-	★	-	★	-
Putten	-	-	-	-	-	-	-	-	★
Tsitsihar	★	★	★	★	-	★	★	-	★
Madrid	★	★	★	★	★	★	★	★	★
Bergamo	-	★	★	★	-	-	★	-	★
St. Luis	★	★	★	★	-	★	-	★	-

[a] References for the studies are given in the main text.

original report. If it is not clear from the original studies whether a certain parameter has been fulfilled, it has not been given a star. We consider a study evaluated by 9 stars to reliably assess all aspects of the NO_x removal efficiency. However, valuable knowledge can still be drawn from all of the studies. In the following, the methodology and main conclusions from each of the assessed studies will be summarized and discussed. The order of the assessed studies is random.

3.1. Field Studies

3.1.1. Antwerp (2004-2010)★★★★★★

Boonen and Beeldens of the Belgian Road Research Center (BRRC) conducted a pilot project in Antwerp, Belgium [57, 58]. A test side of 10.000 m^2photocatalytic pavement blocks containing TiO_2 in the wearing layer (8 mm) was constructed in 2004-2005. The blocks were installed on the parking lanes of the main road in Antwerp. The width of the lanes was 2×4.5 m, with a total road width of 60 m. Initial laboratory testing according to the ISO standard showed an efficiency of approximately 20 % at 30% relative humidity, which was decreasing with increasing humidity, hence reaching approximately 5 % at 70 % humidity. Pavement blocks were taken periodically from the location from 2006 to 2010 and subjected to laboratory testing according to the ISO standard. Good durability is demonstrated over the period of testing. Furthermore, it is shown that rinsing the blocks to remove deposits prior to testing increases the abatement efficiency. *In situ* measuring at the test site was furthermore carried out. NO_x levels were measured three times during the project 5 cm above the ground. Measurements were performed at two test sites with photocatalytically active pavement and a similar reference site. The measurements suggested a decrease in NO_x concentration; however, due to the lack of blank data, Boonen and Beeldens stress that precaution should be taken when interpreting the data.

3.1.2. Guerville (2004) ★★★★★

In a project by Maggos*et al.*, funded by the EU-PICADA project, the abatement efficiency of a TiO_2-containing cement material was tested at a pilot site in the CTG cement plant in Guerville, France [59]. Three artificial canyon streets with dimensions w=2 m, h=5.2 m, l=18.2 m (width to height ratio=0.4) were established. Each canyon was examined for different scenarios. The first street was left untreated as the reference, while the walls of the second street were covered with TiO_2 treated material panels (3% TiO_2 and sand). The walls of the third street were covered with mortar panels not containing TiO_2. A gas emission source was installed to uniformly distribute gas along the walls of the canyon. The NO_x concentration was continuously measured between July 9th and September 3rd 2004, on both sides of the treated and untreated canyons, along with a range of other gas concentrations and meteorological parameters. The pollution source was moved from TiO_2 street to reference street in August 23rd. NO_x concentrations were generally significantly higher in the reference canyon than the TiO_2 coated canyon, with recorded concentrations being 36.7-82.0 % lower in the TiO_2 canyon. The *in situ* measurements were supported by model results, which are not shown in the report.

3.1.3. London (2007-2010) ★★★★★

A field study commissioned by the London Borough of Camden, conducted by Barrat *et al.*, aimed to study the effectiveness of TiO_2 containing paint for NO_xremoval in London [60]. The test site was the 135 m^2 western wall of St. Martin's College, Bloomsbury. The NO_x concentrations were measured at a distance of 0.1 m and 1.5 m from the painted wall. Furthermore, these data were compared with measurements from surrounding monitoring sites in similar urban background situations. Measurements were conducted during three phases. In the co-location phase (47 days from July 2nd to September 4th, 2007), the uncertainty between the two monitoring instruments was assessed. In the pre-intervention phase (16 months from September 4th), monitoring was conducted at 0.1 and 1.5 m from the wall without TiO_2 paint. In the post-intervention phase (March 26^{th,} 2009-January 1^{s}t, 2010), monitoring was conducted after the application of the TiO_2 paint. Interpretation of the results was greatly complicated by the appearance of an unidentified, local source of NO_x interfering with the data at certain wind directions. Therefore, the results were very sensitive to the wind direction and consequently compromising the comparison with the surrounding reference sites. The study found a difference in NO_x concentration at 0.1 and 1.5 m from the wall in the post-intervention phase, with lower NO_x levels closer to the wall; however, similar effects were observed during the nights (*i.e.*, without sunlight). The reason for this observation remains unclear. When the measurements were unaffected by the local source (between 06:00 and 24:00 when the wind direction was North- North-westerly), reductions between 4 % and 7 % in NO_x were observed. After 15 weeks of measurement, the NO_x concentration was suddenly significantly decreased, which might be due to a sudden disappearance of the local source; however, the authors are unable to validate this explanation. Overall, this study underlines the importance of choosing a suitable reference site. In this specific case, the unknown factors influencing the measurements, along with the very small testing area (135m^2), make it hard to draw any meaningful conclusions regarding the effectiveness of the photocatalytic paint.

3.1.4. Rome (2007)★★★★★★★

In a field study conducted by Guerrini, the NO_x abatement efficiency of photocatalytic cement-based coating (Italcementi) in the Umberto I tunnel in Rome was found to be 21 % [61]. The tunnel of dimensions; l= 347.70 m, w=: 17 m, and h= 8.5 m was completely renovated in August 2007. In this context, a new lighting system, including UVA light sources, was installed, and photocatalytic cement-based paint was applied in two layers. The total area covered with photocatalytic paint was 9000 m^2. The NO_x abatement capacity of the paint was assessed according to the NO_x gas recirculation method, adopted by CTG for the quality evaluation of TX active products. In the laboratory tests, a NO_x abatement capacity of 88–90% was observed after 60 min, under UV light irradiation (20 W/m^2). It should be noted that the testing was not executed according to the ISO standard. Before the renovation, measurements were conducted in a reference period from July 3rd, 2007 – July 20th, 2007. Following the renovation, measurements were furthermore conducted in the period from September 25th, 2007 – October 12th, 2007. NO_x monitoring stations were positioned at the sidewalk of each end of the tunnel at 1m height and in the center of the tunnel at heights of 1m and 6m. Furthermore, meteorological parameters and vehicle frequency were measured in the tunnel before and after renovation. The mean hourly value of passing vehicles amounted to approximately 1100 vehicles/hour in both periods of monitoring. The mean NO_x value measured in the tunnel was reduced by 21% (25% for NO and 19% for NO_2). For further comparison, official pollution data from two monitoring stations in Rome were used as an additional reference in the same two periods. At these stations, an increase in NO_x concentration of 61% and 121% were observed from the first to the second period. On this background, Guerrini *et al.* conclude that it is likely that the decreased NO_x concentration in the tunnel is due to the photocatalytic paint. They furthermore suggest that the actual effect might be as high as 64 % taking into account the increase in mean NO_x concentrations at the reference sites. However, these numbers may not be comparable as the reference sites could be affected by various factors increasing or decreasing the NO_x levels.

3.1.5. Hengelo (2009-2010)★★★★★★★★

In a field trial by Ballari*et al.*, Castorweg street in Hengelo, Netherlands, was monitored for NO_x concentrations before and after application of photocatalytic concrete [62]. Prior to the field trial and periodically during the study, the photocatalytic concrete was tested in the laboratory according to the ISO standard. NO_x concentrations and meteorological data were furthermore monitored at the treated section of the street (150 m x 5m) and a non-treated section for reference. The applied concrete blocks consisted of concrete double layer blocks with a 70 mm thick lower layer and an active upper layer of 5 mm with TiO_2. Monitoring stations were positioned at different heights; 5cm, 30 cm, and 150 cm. Following the initial application of photocatalytic concrete blocks in November 2009, no decrease in NO_x values was observed. An additional coating containing 4% TiO_2 was subsequently applied to the blocks in May 2010; however, the durability of this coating was poor, and after 2.5 months, the photocatalytic activity was reduced to the value before the coating. The second layer of coating was applied in September 2010, and after 1.5 months, the activity was still good. After 11 months, the activity was again reduced to pre-treatment levels. During the periods where the photocatalytic layer was supposed to be active, a mean NO_x reduction in the treated section of 19.2 % was observed relative to the reference section. Here, it should be stressed that the authors report a standard deviation of 17.8 %, which is substantial, including only data measured in the afternoon; the reduction was 28.3 %, with a standard deviation of 20.0 %. Overall, this study indicates a positive effect of the photocatalytic treatment; however, the poor durability of the applied product greatly complicated the project. This conclusion furthermore underlines the importance of performing periodic durability testing to ensure continuous catalyst activity during the entire time span of the project.

3.1.6. Malmø (2009-2010)★★★★★★★

In this study, the effect of replacing the ordinary tiles with photocatalytic TiO_2 tiles (from the manufacturer Stark-aBetonindustrier) at the pavement of a highly polluted canyon street in Malmø, Sweden, was tested [63]. The street was divided into two sections, where the tiles were initially replaced with TiO_2 containing tiles in one section while the other section was initially the reference section. Measurements of NO_x concentrations were conducted at both sections in 3 m height from September 26^{th,} 2009 until April 13^{th,} 2010. Subsequently, the tiles in the reference section were also replaced, and additional measurements were made in the period from May 3rdto July 14th, 2010. The overall result of the study was a 5 % reduction in NO_x levels, comparing the difference between NO_x levels in the two sections where only one section of the street was treated. When the data is correlated with wind directions, it was shown that the difference in NO_x levels in some wind directions was as high as 15%.

3.1.7. Manila (2009)★★★★

In this study by Borlaza*et al.*, the NO_x removal efficiency of an ultrafine TiO_2 based paint was evaluated in indoor and outdoor environments in Manila, Philippines [64, 65]. Here, we mainly describe the outdoor experiments. Approximately 6200 m^2 of surfaces along with the highly polluted highway close to the MRT Guadalupe Station in Epifanio Delos Santos Avenue were painted with TiO_2 containing paint. However, some of the painting appears to have been applied inside the MRT Guadalupe Station with no installation of UV light sources. Prior to the painting, measurements were conducted in a blank period of 3 months (December 2008- February 2009). This was followed by an activation period of 2 months (March- July 2009), where the paint was applied, and the actual test period of 4 months (July-October 2009). Sampling was done at roadside stations located along the road and inside the MRT Guadalupe station. Furthermore, one meteorological station was installed to monitor wind speed, relative humidity, and solar radiation intensity. NO_x abatement efficiencies are measured in terms of direct NO_x monitoring (monitoring height or frequency is not reported) and indirectly by monitoring the formation of nitrate using melinex polyester film strips with an area of 80 cm^2 painted with the photocatalytic paint. These were caged in Perspex boxes and placed at one roadside site and one in-station site along with similar paint strips with no photocatalytically active ingredient. These were analyzed periodically by ion chromatography. The direct measurements showed an average NO_2 reduction of 13 % at the roadside sites and 5 % at the in-station sites. This lower reduction is not surprising as UV light intensities are expected to be diminishing at the in-station sites. Large variations in efficiency are observed at the different monitoring sites. Due to the lack of reference data and the long-range of uncertain factors in the study (*e.g.*, monitoring height as well as local meteorological and traffic data for the various monitoring stations), it is hard to draw any reliable conclusions based on the direct measurements. The ion chromatography studies showed a significant increase of nitrate on the photocatalytic paint strips compared to the blank strips at the roadside site. Borlaza*et al.* calculate a total NO_x removal range of 0.285 to 2.242 g/m^2 for 10 months.

3.1.8. Brussels (2011-2013)★★★★★★★★

In a field study by Gallus *et al.*, the effect of photocatalytic cement was investigated in the Leopold II tunnel in Brussels, Belgium [66]. The campaign was initiated in June 2011. After a period of monitoring the untreated tunnel section, the sidewalls and ceiling of a 70 m section of the tunnel were coated with a photocatalytically active mortar (Italcement, TX-Active Skim) in August 2011. Monitoring started again in September 2011. In January 2013, the active section was extended to 160 m, and a more active mortar from the same manufacturer was applied. Two switchable UV light systems were installed (315-420 nm) with intensities of 0.6 ± 0.3 and 1.6 ± 0.8 W/m^2. Due to the preparation procedure, monitoring was not started before 9 and 49 days, respectively, after the application of the mortar. Two monitoring stations were installed at each end of the tunnel, upwind and downwind of the active section. Sampling was performed 33 and 20 cm below the ceiling from June-September 2011 and in 34 and 44 cm in January 2013. To minimize the influence of fluctuations in atmospheric composition and traffic flow inside the tunnel, NO_x concentrations were normalized to the CO_2 concentration, which was assumed to be unaltered by TiO_2 and emitted by the cars along with the NO_x gasses. Besides the field measurements, the possible deactivation of the photocatalyst was studied in the laboratory according to a modified ISO approach, simulating the tunnel conditions. Three different approaches were applied in order to compare the active surface with an inactive surface under similar conditions: *i)*NO_x levels were measured before and after application of the active coating, *ii)* NO_x levels were measured upwind and downwind of the active tunnel section with the lamps on *iii)* NO_x levels were measured at the downwind site with the UV radiation on and off. Sub-conclusions from each approach were: *i)* No significant NO_x remediation was observed, as before and after measurements (both campaigns) were indistinguishable within the uncertainty limits (two standard deviations). *ii*) The upwind and downwind NO_x/CO_2 ratios were likewise indistinguishable within the uncertainty levels in both campaigns. When looking only at low wind speeds where increased photocatalytic activity is expected, similarly, no significant effect was observed. The authors estimate an upper limit of pollutant reduction of 3% based on error propagation of the NO_x/CO_2 ratio and the precision of the two instruments used. *iii)* Comparing the NO_x levels measured when the UV light systems are turned on with those when they are turned off also shows no significant NO_x remediation.A 2 % remediation is estimated following the same procedure as in *ii)*. Taking into account the results of the three approaches, it is highly unlikely that the low reduction in NO_x observed is due to an increase in emission after the pre-campaign. Gallus *et al.* suggest that the low activity of the photocatalytic coating is due to deactivation caused by the low UVA irradiance in the tunnel (requested value is 4 W/m^2), as well as accumulation of particles and organic grime from the dirty tunnel blocking the active sites. Since the active monitoring campaign did not include the freshly coated surface, the surface might have been covered in pollution already at the beginning of the campaign, which would explain the low activity. Gallus *et al.* also conduct an upper limit calculation assuming first-order uptake kinetics, which yields a maximum theoretical reduction of 20 % at optimum conditions.

3.1.9. Louisiana (2010-2011)★★★★★

Hassan *et al.* conducted a laboratory and field study to investigate the photocatalytic activity of asphalt pavements on the campus of Louisiana State University [67]. Asphalt mix substrates were coated with a water-based surface coating TiO_2 spray (2%vol) and subjected to laboratory testing according to a modified JIS TR Z 0018 standard. The durability of the samples was furthermore tested in an accelerated loading test (ALT), mimicking very rough wearing conditions. The laboratory tests showed a NO_x removal efficiency of 31-55%. It should be noted that these numbers are not directly comparable to other studies as they were not obtained according to the ISO standard. The ALT resulted in a significant reduction in removal efficiency of 68%. The effect of various environmental factors on the NO_x removal efficiency was studied, and the results are in agreement with other studies [37]; efficiency is decreased with increasing humidity, increasing wind speeds, and decreasing UV intensity. In the field study, the existing asphalt at a pavement site at Louisiana State University was sprayed with the TiO_2 surface coating. Traffic count, meteorological parameters, and NO, NO_2 and NO_x concentrations were monitored onsite. NO_x concentrations were measured directly and indirectly (accumulated nitrate). The same monitoring station was moved between a coated and an uncoated area. Further details on the location of the coated and uncoated areas are not stated in the paper by Hassan *et al.* The data were furthermore compared with measurements from a nearby monitoring station. It was not reported whether conditions at this site were comparable to those at the test site. Monitoring data collected 10 days before and 10 days after the application were directly compared to assess the NO_x reduction efficiency. NO_x levels in the period after application were generally lower than those before. Likewise, NO_x concentrations in the treated section are lower than those in the non-treated section. Even though the data suggest a photocatalytic activity, one should be careful drawing conclusions, as differences in traffic and weather conditions are not considered which make the comparison hard, in particular due to the short duration of the campaign.

3.1.10. Wijnegem (2011-2013)★★★

As part of the ECO2PROFIT project, precast concrete with TiO_2 in the wear layer was tested in Wijnegem, Belgium, by Boonen and Beeldens [58]. Initially, extensive laboratory studies were performed to optimize the concrete with respect to photocatalytic activity without altering other properties of the concrete. The most significant result from the laboratory tests was that the application of a curing compound to the young concrete was found to inhibit photocatalysis. Hence Boonen and Beeldens recommended that photocatalytic spray coating is not applied prior to some months after the curing compound. Concrete pavement was constructed between May 15^{th} and 18^{th}, 2011, in the industrial zone in Wijnegem. The top layer was commercially available (CBR, Heidelberg Cement Group) white cement containing 4% TiO_2. Some parts of the road were treated with a curing agent, while some were instead covered with a plastic sheet. An *in situ* ISO test setup was used to periodically measure the activity of the concrete. In agreement with the laboratory experiments, the curing agent was initially found to inhibit the activity; however, this effect decreased over time. *In situ* ISO test data were collected after 5, 17, and 29 months, and the concrete was still active after 29 months. No measurements of the effect on the air quality were performed.

3.1.11. Gasværksvej, CPH (2012-13)★★★★★★★★

In a field study by Folli *et al.* from 2015, photocatalytic paving blocks were tested in a canyon street, Gasværksvej, in Copenhagen, Denmark [68]. The test area consisted of a 200 m long street section, 100 m with photocatalytic pavement blocks on both sides, and 100 m with ordinary pavement blocks. The blocks were installed in June 2012, and the monitoring campaign ran from April 2012 to August 2013 (blank test from September 2011 to April 2012). NO_x levels were monitored at the test side and the reference side 2 m above the ground along with temperature and relative humidity. Measurements were taken every minute throughout the entire duration of the campaign. Prior to the field study, laboratory tests showed a photocatalytic activity of 78% (calculated average pseudo surface reaction rate is 4.09 x 10^{-4} ml s^{-1} m^{-2}). It is not specified whether this activity is obtained according to the ISO standard. The field experiments showed a significant decrease in NO_x concentrations, along with no major difference in NO_2 levels comparing the active site with the reference site. The efficiency was highest at the summer solstice, where a monthly abatement of 22% was observed.

The results were later criticized in a comment by Kleffmann [69]. Kleffmann showed that assuming canyon parameters of 100m ×15m ×15m giving an active surface area to volume ratio of 0.0178 m^{-1}, and first-order reaction kinetics, an upper limit photocatalytic NO uptake of 0.8% was obtained. Whereas this is definitely a relevant number, it may not be directly comparable with the results from the real-life test site, as monitoring was done locally 2 m above the treated pavement. Significantly lower reductions are to be expected considering a box of the dimensions assumed by Kleffmann. A theoretical upper limit of similar magnitude is obtained in the same comment considering the flux considerations, taking into account all of the NO_x emitted by vehicles in the street. Likewise, this number may not be comparable with the locally measured reductions by Folli *et al.* Kleffmann also questions the explanation given by Folli *et al.* that why no NO_2 reduction is observed, as NO and NO_2 are closely related in tropospheric chemistry *via* the Leighton mechanism, in which NO and NO_2 are rapidly converted back and forth on timescales of less than a minute [70]. Kleffmann suggests that the freshly emitted NO gas downwind to the measuring sites are transported to the reference site first (where the Leighton equilibrium has not been established yet), and the NO_2 concentrations are increased when the air reaches the test site. Kleffmann recommends that photocatalytic street canyon studies should be accompanied by canyon modelling to avoid overestimation of the impact.

3.1.12. Copenhagen Airport (2013)★★★★★★★

In a field campaign from 2013 by Jensen and Pedersen, the effect of NO_x removing concrete (Photocat NOxOFF) was tested [71]. The initial activity of the concrete was tested in the laboratory according to the ISO standard. The field test site was a parking lot at Copenhagen Airport (kiss-and-fly). The parking lot was divided into a reference section of approximately 250 m^2 and an active section of approximately 250 m^2. NO_x levels were monitored 0.5 m above the ground at both sites 41 days before installation (March 17^{th} to April 28^{th}) and 43 days after installation (May 11^{th} to July 4^{th}). Considering the relative difference between the reference site and the active site, an average decrease of 13 % in NO_x is observed, including both daylight and dark hours, where no photocatalytic activity is expected. Meteorological data was not monitored.

3.1.13. Konnigstunnel- The Hague (2013)★★★★

A colloidal photocatalytic ultrafine TiO_2 product (10 %) was evaluated in the lab and a field study in the Konnigstunnel in The Hague, Netherlands [72]. The laboratory testing was conducted according to the ISO standard; however, the results were not reported. Photocatalytic paint was applied directly to the walls and ceiling on a trial section of 150 m of the 650 m long tunnel. NO_x concentrations were measured by chemiluminescence. Additionally, CO_2 and ozone concentrations, as well as meteorological parameters and traffic counts, were monitored. UV light intensities in the tunnel were 1 W m^{-2} (walls) and 0.6 W m^{-2} (ceiling). The lights were switched on and off in periods of 24 hours as an additional reference. Blank tests were performed for a short period of time before application. The duration of the test period is not reported. The authors claim that their chemiluminescence measurements indicate a reduction in NO of about 20%. It is not reported how this number is obtained. The result is supported by a mathematical model (which is not further explained) yielding a theoretical maximum removal efficiency between 10 % and 55 %. Nitrate accumulation measurements were furthermore conducted in an attempt to eliminate the problems involved in measuring NO_2 by chemiluminescence. Paint strips were placed in the beginning and end of the trial section at two positions (close to the ceiling and 1 m above the floor). It is not reported when and how many times these were collected. More nitrate was generally accumulated at the ceiling, which is explained by the excess water from traffic at the lower position, possibly washing out the nitrate. Also, more nitrate is accumulated at the beginning of the trial section; however, no attempt to explain this is made. The results clearly illustrate that the collection of accumulated nitrate also presents challenges as a NO_x reduction measure.

3.1.14. Fælledvej (2014)★★★★★★

As part of the Light2Cat EU project, 125 m of paving tiles containing a TiO_2photocatalyst (Starka) were installed at both sides of a road, Fælledvej in Copenhagen, Denmark, from August 2014 to October 2014 [73]. NO_x levels were monitored 1.5 m above the surface of the tiles. Furthermore, a weather station was installed monitoring meteorological parameters. The trial campaign ran from October 2014 to June 2015 and was compared to a blank period from October 2013 to June 2014. Measurements were taken every minute throughout the campaign. The study concluded a NO_x reduction of 9.6-19.9% depending on the light intensity. Surprisingly, the most significant reductions were observed for NO_2. Most notably, NO reductions at light intensities >200 and >300 W/m^2 were 0.7 % and -6.6 %, respectively, while the same numbers for NO_2 were 18.7 % and 17.7 %. Also, it is unphysical that the efficiency appears to decrease with increasing light intensity. As the measuring technique and the number of data points collected at each solar radiation intensity are not reported, it is not possible to conclude whether these numbers are describing a real trend or if they are not statistically significant or caused by, *e.g.*, calibration issues.

3.1.15. Holbækmotorvejen (2015)★★★★

The Light2Cat EU project also included a field study of photocatalytic jersey barriers at Holbækmotorvejen, Denmark, a highly trafficked highway [73]. The test site consisted of an active site with three photocatalytic jersey barriers and a similar reference site with regular jersey barriers. At both sites, one of the barriers was caged in a Stevenson screen in order to simplify monitoring. NO_x concentrations were measured inside the screen. Furthermore, a weather station was installed monitoring meteorological parameters. Monitoring was conducted during two different periods of time; March 2015-April 2015 and July 2015-August 2015. Data points were collected each minute throughout the two campaigns. For the first monitoring campaign, no significant NO_x abatement was observed except at light intensities above 600 W/m^2, for which a reduction of 9.8% was observed compared to the reference site. For the second period, efficiencies ranged from 5.6 (>10 W/m^2) to 7.9% (>600 W/m^2) were observed. The authors stress that the actual abatement efficiencies are expected to be higher, as the presence of the Stevenson screen is likely to inhibit washing away of accumulated nitrates blocking the active sites.

3.1.16. Valencia (2015)★★★

The third part of the Light2Cat project was a field study in Spain, where a total of 535 m^2 of standard paving tiles at Calle Felix Pizcueta were replaced by photocatalytic tiles (Hermo) [73]. NO_x concentrations were monitored at 1.5 m above the surface, and meteorological parameters were not measured. Reference data were obtained from November 2014 to January 2015, while the trial data were obtained during the period from April 2015 to June 2015. The study shows a substantial decrease in NO_x concentrations; however, no conclusion on the efficiency of the photocatalytic pavement can be drawn, in part due to the reference period being a period where lower light intensities, hence higher NO_x concentrations are expected. Even more importantly, dramatic changes in the traffic took place between the blank period and the active period due to construction work. Hence the data will not be discussed in further detail here.

3.1.17. Roskilde (2014-2015)★★★★

The trial study was conducted by Photocat A/S in cooperation with the municipality of Roskilde in Denmark [71, 74]. Two parking lots of a total area of 5.000 m^2 were activated by treatment of the existing asphalt with Photocat'$sTiO_2$ based NOxOFF technology. The activity and adhesion of the photocatalytic liquid applied to the parking lots were initially tested in the laboratory following the ISO-22197-1 standard, and the durability was tested according to an accelerated aging test according to the EN 1297-2004 standard. The accelerated aging laboratory test results showed a decrease in NO_x activity of 17 % after conducting an accelerated duration test corresponding to 2 years in real-time. Both locations were treated on September 29th, 2014. An *in situ* ISO test setup was developed to periodically monitor the activity of the asphalt. The activities were measured at ambient and artificial conditions. Measurements were taken on three separate days over a period of two years. After the two years measuring period, the activity was not significantly decreased.

3.1.18. Toronto (2017-2018)★★

The construction of a highway sound barrier treated with a photocatlytically active coating SmogStop^{TM} was completed in March 2017 [75]. On August 18th, 2017, data collection began and continued until February 28th, 2018. No initial laboratory tests ensuring the activity of the product were reported. Chemiluminescence analyzers were installed on-site to monitor NO_x concentrations. The barrier consisted of three bays, of which only the middle one was coated. Air was sampled at the inlet and outlet of the treated section of the barrier. The inlet data was then used as a reference, while the outlet data, where the air had passed over the treated section, were considered as the treated sample. It is not clear at what distance from the barrier the sampling was conducted; however, it appears to be in very close proximity to the surface. Blank data were not collected before the installation of the photocatalytically active barrier. Wind data were furthermore collected. Data were collected at 1-minute logging intervals using a 10 second time constant. Observed average monthly daytime NO_x reductions vary from 21% (November) to 54% (December), with maximum daytime reductions reaching as much as 92%. The average reduction of the study was reported to be 34%. Average nighttime reductions, caused by the artificial light of the highway and claimed visible light activity of the photocatalytic material, were claimed to vary from 22% to 44%. The results obtained in this study should be viewed as an *in situ* activity test rather than an actual study of the effect on the ambient air quality due to the lack of an untreated barrier for reference, blank data as well as the supposedly short monitoring distance.

3.1.19. Putten★

As part of the Dutch Air Quality Innovation Program (IPL), the effect of noise barriers with photocatalytic surfaces was tested in the Netherlands [76]. Four panels with different coatings were fitted onto an existing noise barrier along with the A1 at Terschuur. Later, a porous noise barrier optimized for air quality with a photocatalytic coating was constructed along the A28 at Putten. Unfortunately, very limited data of the study is available [76]. The IPL report states that NO_x concentrations were measured along with meteorological parameters, but no information was given about the duration of the project, blank tests, or reference data. The report finds no evidence for improvement of the air quality, which is explained by the short contact time between air and barrier along with unfavorable meteorological conditions.

3.1.20. Tsitsihar (2011)★★★★★★★

In a study by Chen *et al.*, a stretch of road at the Zhonge Toll Station on federal highway G11, China, was divided into two sections (each of approximately 1600 m^2), where one was sprayed with a TiO_2 containing coating, while the other one was left with no coating, [43]. Air sampling at the two sites was performed simultaneously during a period of 3 months. The sampling method involved the absorption of contaminants onto filters containing N-(1-naphtyl)-ethylenediamine followed by colorimetric determination of NO_x concentrations. A total of 18 monitoring sites were placed at both sites, and air sampling was performed at the height of 0.5 m. No blank test was performed. Meteorological parameters, as well as traffic counts, were monitored continuously. The study showed decreased NO_x levels at the coated site compared to the reference site, with the effect being dependent on light intensity. The outdoor test study was supported by laboratory results showing abatement efficiencies of 78.2 % for NO and 58.5 % for NO_2; however, these results were not obtained according to the applicable ISO standard. The activated concrete was furthermore subjected to a polishing experiment to test the durability of the product. It was found that the NO and NO_2 abatement efficiencies after artificial wearing was 37.4 % and 25.8 %, respectively, under lab conditions comparable to the initial tests.

3.1.21. Madrid (2014-19) ★★★★★★★★

The European-funded life project, LIFE-PHOTOSCALING, performed in Madrid, combined real-scale testing with the to-date most comprehensive initial testing program and data analysis [77]. The project was started in October 2014 and ended June 30, 2019. Prior to the real-life scale campaign, extremely comprehensive laboratory and in-situ testing were performed, resulting in the development of an indicator-based framework for assessment of the performance and conformity of photocatalytic pavement materials [78-81]. *In situ* testing was performed at two test sites in Madrid, each 900 m^2, consisting of several 32 m^2 workbenches. In each platform, three different kinds of slabs were tested; asphalt covered with photocatalytic slurry, asphalt, and urban tiles, and finally, concrete supporting photocatalytic tiles (precast concrete and ceramic tiles). The workbenches were furthermore compared to non-photocatalytic workbenches of identical composition. At each site, meteorological parameters were monitored. Alongside the in-situ tests, laboratory tests were performed addressing intrinsic performance, photocatalytic performance, and undesired side effects, including nitrate selectivity and Ti leaching. The project developed and submitted a patent for a new way of testing and evaluating photocatalytic surfaces *in situ* with a PHOTOSITE device [79]. The NO_x removal ability of PHOTONSITE was validated by comparison with the standard ISO 22197:1-2007. Based on the laboratory and in-situ performance testing, an indicator-based decision tool to assess the sustainability of a particular material was developed, the *Photoscaling decision-maker*, which is publically available online (https://www.life-photoscaling.eu/decisionmaker/). The three indicators were photocatalytic efficiency, intrinsic performance, and undesired secondary effects, divided into 4, 1, and 3 sub-indicators, respectively. The authors propose a scoring system enabling objective comparison of the performance of different products. The decision-maker furthermore includes a life cycle analysis (LCA) and estimation of the NO_2 removal percentage estimated from a statistical model based on data from the PHOTOSCALING platforms. Using the highest scoring product from the initial testing, photocatalytic emulsion was applied to 4200 m^2 of pavement of a selected street, including sidewalks, parking area and asphalt. A similar street was selected as the reference site. The application was performed in December 2018. Samples were taken before and 3 times during the campaign to test the material according to the before mentioned criteria. Monitoring was performed by 42 and 33 different censors placed in different heights in the two streets, respectively to account for vertical concentration gradients. The relative difference between the two streets were monitored in the same months before and after application (Feb to May 2018 and 2019). From the first three months of monitoring, the study estimates an average NO_2 removal efficiency at ground level of 28 %.

3.1.22. Bergamo (2006-2007)★★★★★

The project in Bergamo included a renovation project for an about 500 m long road and sidewalk with a total of 12,000 m^2 of photocatalytic material [82]. The renovation project was completed in September 2006, and two environmental monitoring campaigns of each 12 days were conducted in November 2006 and January 2007. Measurements were taken on two different sites, a reference site of bituminous concrete without photocatalytic material and the area where the photocatalytic blocks were installed. Initial measurements prior to the application were not performed to ensure comparability of the two sites. During the first measuring campaign, two NO_x chemiluminescence analyzers were used, and the height of the sampling was approximately 1 m. During the second campaign, 4 chemiluminescence analyzers were used simultaneously at two different heights – 30 and 180 cm. In addition, meteorological data like temperature, wind speed, and atmospheric precipitations were recorded together with a number of vehicles traveling during the daytime at the site. The results from the two testing campaigns highlighted efficient abatement of pollutants by the photocatalytic pavement compared to the reference asphalt pavement site. During the nighttime, the two control stations recorded very similar NO_x values. Evaluating the NO_x data from different heights showed higher NO_x reduction near the ground (30 cm) compared to at 180 cm. NO_x reduction of appr. 30 % were reported at 30 cm and 20 % at 180 cm; however, these numbers should be considered uncertain due to the lack of blank data. The study did not include laboratory data or continuous durability measurements.

3.1.23. St. Luis (2011-2012)★★★★★★

The St. Luis field study presented by Sikkema [47] was constructed using TX Active cement on a project at the Missouri Route 141 in the western St. Louis metropolitan region. The selected site was part of a newly constructed alignment of Route 141. 1500 ft test section of photocatalytic concrete pavement (38ft in width resulting in 57,000 ft^2 or 5,300 m^2) and 1500 ft of conventional concrete pavement were situated on the southbound lanes. Four monitoring methods were used to assess NO_x, including passive ambient air monitoring and laboratory evaluation of fresh and aged samples. Using a passive sampler (Ogawa), no significantly lower NO_x values were observed on the photocatalytic pavement compared to the control pavement. Laboratory durability studies found a 48 % decrease in the NO_x removal rate comparing fresh and aged mortal slabs following ISO 22197-1. The study expresses concern about non even traffic behavior on the reference site *vs*. the photocatalytic site (variable vehicle speed due to highway entrance on one of the sites only) which could lead to different NO_x emissions and since no blank measurements were made before installing the photocatalytic pavement, it is not possible to predict whether the NO_x levels were the same on both sites before starting the experiments which make it difficult to predict the NO_x mitigation from the photocatalytic pavement. Passive samplers were the primary monitoring devices to measure the air quality of the project. Ogawa passive samplers were used to collect NO_x data. 3 samplers were installed at 30 cm above the pavement and 3 samplers at 100 cm above the pavement. The samplers were set in place for 14 to 35 days period before being replaced with fresh sampling discs. The drawback of using passive samplers is the poor time resolution (weeks or months). From passive samplers, it is not possible to investigate the photocatalytic effect as a function of, *e.g*., sun light and thereby retrieve important information in night time where there is no light and thereby no photocatalytic effect. One 20 hours monitoring campaign with a 2B technologies monitor was presented. From this monitoring campaign, it is evident that during nighttime, the NO_x of the reference is similar to the active site, whereas, during the daytime, the difference is up to twice the concentration at the active site compared to the reference site.

3.2. Discussion

Designing a field test-study is far from straightforward due to the many variables influencing the experiment and the expenses related to the execution of a large-scale study of long duration. Of the 23 studies assessed here, only the Madrid study fulfills all of the criteria, and we consider it to provide valuable information for test evaluation. Generally, two different types of test studies have been performed up till now. 1) Periodic activity measurements performed either in the laboratory or *in situ* can be used to determine the durability of a photocatalytic material but cannot be used to directly quantify the potential of improving ambient air quality. This type of experiment was done in the Antwerp, Hengelo, Louisiana, Brussels, Wijnegem, Roskilde, Tsitsihar, Madrid, and St. Luis studies. The studies in Hengelo and Brussels clearly demonstrated the importance of periodically testing the activity to avoid concluding a low effect of the material that is actually caused by the photocatalyst no longer being present at the surface.2) Monitoring of ambient NO_x levels at a test site where a photocatalytic surface is installed to quantify the improvement of the air quality. This kind of experiment has been attempted in Guerville, London, Rome, Hengelo, Malmø, Manila, Brussels, Louisiana, Gasværksvej (Copenhagen), Copenhagen Airport, The Hague, Fælledvej (Copenhagen), Holbækmotorvejen (Copenhagen), Valencia, Toronto, Putten, Tsitsihar, Madrid, Bergamo, and St. Luis. While the first type of experiment is relatively straightforward, the second type of experiment is associated with a high level of uncertainties and should therefore be designed with precaution. The most common shortcoming, which we also consider the most fatal, is the lack of reliable reference data and blank data. The combination of test data, reference data, and blank data minimize influences from changes in NO_x levels over time as well as local differences. Of the 20 studies measuring ambient NO_x levels, only seven studies, *i.e*., Hengelo, Rome, Malmø, Brussels, Gasværksvej, Copenhagen Airport, and Madrid, present both. The area of the photocatalytic surface is another crucial parameter when designing a test site. The smaller the area, the more sensitivity to uncertainties and local fluctuations is to be expected. Furthermore, a small active area can only be expected to result in very local reductions of detectable magnitude. There are, to the best of our knowledge, no official recommendations to the minimum size of a photocatalytic test surface. Here, we adopted a generous limit of 250 m^2 to exclude only very small areas. Using a larger area would expectedly lead to more reliable data collection.

CONCLUSION

TiO_2 based photocatalysis has the ability to remove hazardous air pollutants, *e.g*., NO_x. The incorporation of this technology into surfaces in urban areas, such as pavements, asphalt, tunnel walls, *etc*., is a promising tool to improve air quality at a low cost in areas where pollution levels, as well as population densities, are high. Quantifying the actual reductions in real life is challenging due to the many parameters affecting the test results, such as weather conditions and traffic level, and real-life studies with conflicting results have been presented in the literature. In this review, we have attempted to evaluate 23 existing field studies based on the information available in the publications against nine specific criteria. We furthermore summarized the main conclusions that can be drawn from each study. The evaluation criteria should serve as an aid to interpret existing results, as well as guidelines for future test study designing; however, they should not be viewed as a ranking of the studies. We recommend that future *in situ* studies present monitoring results from a blank period prior to construction of the photocatalytic surface, reference data from an inactive site closely resembling the active site, initial laboratory test results of the photocatalytic activity, periodic activity test results to ensure the durability of the product as well as supplementary meteorological data, *etc*. We furthermore recommend that the area of the active site should be at least several hundred of square meters, while the duration of the project should be on the time scale of months. The monitoring distance should be clearly stated in the report and should ideally represent human exposure height.

CONSENT FOR PUBLICATION

Not applicable.

FUNDING

None.

CONFLICT OF INTEREST

The authors declare no conflict of interest, financial or otherwise.

ACKNOWLEDGEMENTS

Declared none.

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