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RESEARCH ARTICLE

In situ investigation of NO_x photocatalytic degradation: Case study in an open space office in Manchester, UK

Julie Hot[1*] Paul Bradley[2] Jayson Cooper[2] Barnabé Wayser[3] Erick Ringot[1,4]

Abstract: Indoor air is contaminated by numerous pollutants, which impact human health, comfort and productivity. These pollutants have various indoor sources such as building materials, furniture, combustion appliances or tobacco smoke. However, the pollution also comes from outside. In urban area, nitrogen oxides (NO_x) emitted into the atmosphere can reach alarming levels. These traffic-related pollutants, which seriously impact the global environment and human health, can infiltrate inside buildings. Therefore, limiting the amount of breathable NO_x in outdoor and indoor environments is an important priority for the modern society. The photocatalytic process has attracted particular attention in the last two decades and has proved to be efficient to reduce the concentration of NO_x. However, further work has to be conducted to assess its efficiency in real indoor environments. The purpose of this paper was to report on the indoor air quality in an open space office in Manchester, UK. Focus was made on nitric oxide (NO) and nitrogen dioxide (NO_2). The indoor concentrations of both gases were monitored from 14 January 2019 to 7 April 2019. During this period, a photocatalytic coating was applied to a part of the indoor wall. The influence of this coating on the level of NO_x was assessed by comparing the indoor concentrations before and after the application. An attention was paid to the correlation between outdoor and indoor pollution and to the effect of other parameters such as temperature, humidity, pressure and ozone (O_3) concentration. The results showed that the photocatalytic process led to a decrease in the NO_x concentration. The likelihood to find concentrations above 35 ppb for NO and 7.5 ppb for NO_2 was clearly reduced after the coating application.

Keywords: nitrogen oxides, in situ, office, photocatalysis, titanium dioxide, indoor air quality

1 Introduction

Indoor air quality (IAQ) has become a real concern as it seriously impacts human health, comfort and productivity. Unlike outdoor air pollution, interior air pollution remained relatively unexplored until the early 2000s. In 2006, the World Health Organization started to define guidelines for IAQ[1] and identified the three most problematic indoor air pollutants for public health: biological indoor air pollutants[2]; chemical indoor air pollutants[3] and pollutants from indoor combustion of fuels[4]. Building materials, decorative products and furniture are regularly mentioned as potential sources of chemical pollution in indoor environment due to their rather strong emissions of volatile or semi-volatile substances and large surface area[5–7]. Building dampness can also reduce the quality of life of occupants as it leads to an increased exposure to various types of microorganisms (including moulds and bacteria), which emit microbial volatile organic compounds when growing on building materials[8,9]. Moreover, IAQ is threatened by outdoor pollutants. In urban areas, nitrogen oxides (NO_x) and benzene, for example, are serious pollutants, which can infiltrate inside buildings[10–12].

NO_x is primarily made up of two pollutants, namely nitric oxide (NO) and nitrogen dioxide (NO_2). All combustion processes produce NO_x. Indoor sources are generally cooking, heating and smoking, and road transport is the main NO_x outdoor source[13,14]. Guideline values (indoor and outdoor) are only defined for NO_2 due to its serious impact on health: 200 μg/m^3 for short-term exposure and 40 μg/m^3 for long-term exposure[3,15]. However, as NO easily converts to NO_2 in the air, it is of utmost importance to limit emissions of both NO and NO_2. In this end, preventive measures, which principally concern

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* **Correspondence to:** Julie Hot, LMDC, INSA/UPS Génie Civil, 135 Avenue de Rangueil, 31077 Toulouse Cedex 4, France; Email: hot@insa-toulouse.fr
1 LMDC, INSA/UPS Génie Civil, 135 Avenue de Rangueil, 31077 Toulouse Cedex 4, France
2 Guard Industry, Office 2, Chipstead Road, Birmingham B23 5HD, United Kingdom
3 Guard Industrie, 7 Rue Gutenberg, 93108 Montreuil, France
4 LRVision SARL, 13 Rue du Développement, 31320 Castanet-Tolosan France

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the transport sector, have been taken by policies[16–18]. In London, the reduction of traffic emissions is one of the main concerns of the air quality strategy[19,20]. For example, an Ultra Low Emission Zone (ULEZ) operates since April 2019 in Central London to help to improve air quality. The same is true in France, where other actions such as grants for electrical vehicles and speed limit reduction have been taken[21].

In addition to these actions, which focus on outdoor pollution, remedial measures could be implemented to further improve IAQ, combat gaseous pollutants, such as NO_x coming from outside, and reduce health effects related to air pollution. Indeed, it has to be noted that a lower IAQ guideline based exclusively on health criteria was proposed by the French organization ANSES: 20 $\mu g/m^3$ for long-term exposure[22]. The photocatalysis is one of the promising techniques for air purification. This process, widely documented in the literature, is based on a semi-conductor activation, TiO_2 being the most famous. Under irradiation, reactive species (HO, O_2 and HO_2 radicals) are created at the surface and react with pollutants to decompose or mineralize them through redox reactions[23]. The application of TiO_2-based coatings to building materials to degrade NO_x and volatile organic compounds (VOCs) has received increasing attention in recent years and was notably studied in[24–27]. However, most of research work in the literature evaluated their efficiency at lab scale under controlled conditions, which are not representative of the real environment.

This paper focuses on the NO_x indoor concentration in an open space office located in the city centre of Manchester, UK. Maintaining good IAQ in working places is indeed essential to assure health and productivity of employees. It presents a real case study, whose objective was twofold: (i) to collect data on indoor levels of NO and NO_2 in offices and compare them to outdoor concentrations; and (ii) to provide information on the efficiency of a photocatalytic coating (DToxGuard® Int) to reduce NO_x in an indoor environment surrounded by a highly road traffic area. It was led in collaboration with Guard Industry (Birmingham, UK), LRVision (Castanet-Tolosan, France), and the Laboratory for Materials and Durability of Construction (LMDC, University of Toulouse, France). The NO_x concentration was monitored before and after the application of the coating to a small part of a wall. The results reveal a decrease both in NO and NO_2 concentrations, which means promising perspectives for IAQ.

2 Materials and Methods

2.1 Air quality monitor

The monitoring of the IAQ was carried out with the AQMesh® station, which is a small-sensor air quality monitor manufactured in the UK (Environmental Instruments Ltd, Stratford-upon-Avon). According to the manufacturer, it has been designed to offer an easy-to-use and reliable air quality monitoring system that can deliver real-time localized data and analysis. This kind of device proves to be very useful in gathering air quality data in order to support initiatives to reduce air pollution and its risk to human health.

The AQMesh® monitor was configured for measuring NO, NO_2 and ozone (O_3) concentrations in addition to temperature, humidity and atmospheric pressure. One reading was recorded every minute. More than 120,000 readings were collected during this monitoring campaign, from 14 January 2019 to 7 April 2019. This time period included the pre-air quality monitoring (from 14 January 5 pm to 4 March 2019 midnight) before the coating application. The aim was to gather background data and have a reference in order to assess the influence of the DToxGuard® Int product on the NO_x concentration (from 6 March to 7 April 2019 midnight).

2.2 Site description

The monitoring campaign was conducted in Sir Robert M^{c}Alpine (SRM) office in Manchester, UK. Figure 1 shows pictures of the open space office, in which the AQMesh® air quality monitor was placed on the wall, on which the photocatalytic coating was applied.

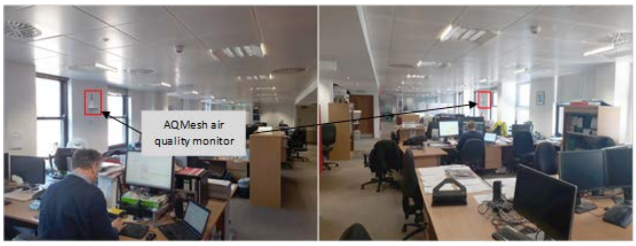

Figure 1. Interior views of Sir Robert M^{c}Alpine office. The AQMesh® air quality monitor was placed on the wall between two windows

Before starting the monitoring campaign, information on site location, surrounding area and open space office characteristics were gathered. They are summarized in Table 1. The site of investigation was in the city centre of Manchester, an area of heavy traffic. Figure 2 shows the location of the SRM building, which is surrounded by two automatic air monitoring stations managed by the Manchester City Council.

2.3 Photocatalytic coating and application

Guard Industry and LRVision compagnies have been working on the air quality issue since 2009. They own a French patent on water based photocatalytic coatings

Table 1. Information on site location, surrounding area and office investigated

Site location	Company	Sir Robert M^{c}Alpine, a building and civil engineering company
	Country	England, UK
	Region	North-West England
	City	Manchester M2 3WQ
	Street	15 Oxford Court
	Floor	3rd
Surrounding area	Traffic	City centre traffic (close to the A57 Manchester Road)
	Airport proximity	13.12 km ~ 18.15 miles
	Air quality information for Greater Manchester	Clean Air Greater Manchester https://cleanairgm.com
Open space office characteristics	Ceiling height	~ 2.8 m
	Surface	400 m^2
	Facade orientation	South-West
	Mechanical ventilation	Yes (no flow rate data)
	Averaged number of occupants	Not provided (no smoker allowed)
	Hours of occupation	From 7 am to 6 pm
	Nature of the joineries (material, age, mechanism)	Wood, 20 years old, vertical sliding
	Nature of the bulbs	Sylvania T5 FHE Luxline Plus 14W 840 | 55cm - Cool White

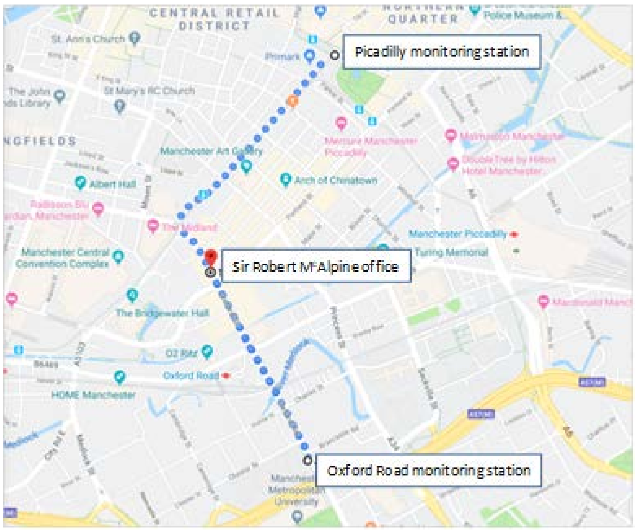

Figure 2. Google map of Manchester city centre. The locations of SRM building and of two air monitoring stations managed by the Manchester City Council are specified

and have investigated the air depollution application of the photocatalysis in collaboration with the LMDC. DToxGuard® is an innovative air purifying product based on the Guard Industry and LRVision patented photocatalytic technology. It was introduced into the marketplace in 2018 and has two variants: DToxGuard® Int (internal) and DToxGuard® Ext (external). The photocatalysis activation occurs when the treated surface reacts thanks to natural and artificial UV light.

DToxGuard® Int is a two-component product that was carefully mixed before use. After mixing it consists of a photocatalytic dispersion containing 6 wt% of dry matter TiO_2. Then, to ensure the application was controlled, DToxGuard® Int was applied by brush (2 layers) and windows were opened to provide adequate ventilation (Figure 3(a)). For this experiment, the treated wall surface consisting of painted plaster was only 2.88 m^2, which corresponded to the area of wall where the AQMesh® air monitor was hung on, from window to window, and floor to ceiling. Once the application was completed, the appearance of the substrate remained unchanged (Figure 3(b)). The wall was coated on 5 March 2019.

Figure 3. (a) Application of DToxGuard® Int by brush and windows opened; (b) After completion: the AQMesh® air quality monitor was setup on the treated wall

Moreover, to keep SRM employees informed of the process, Guard Industry created the sign showed in Figure 4 to give a simplistic explanation of how DToxGuard® Int works.

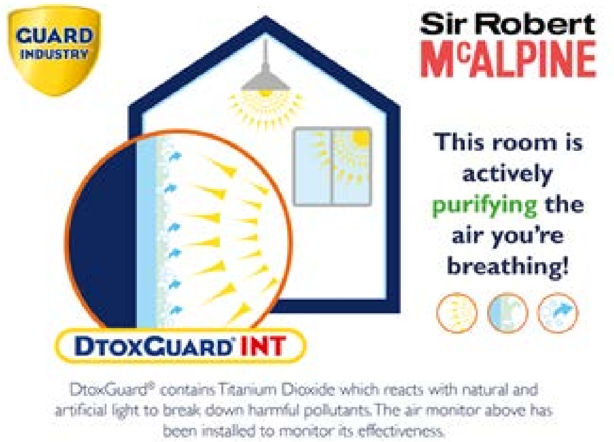

Figure 4. Sign explaining the DToxGuard® Int action intended to SRM employees

3 Results

3.1 NO and NO_2 indoor concentrations

The evolutions of NO and NO_2 indoor concentrations as a function of time are shown in Figure 5(a), 5(b) respectively. The DtoxGuard® Int coating was applied to the wall on the 5^{th} of March 2019.

These data highlight the significant variations of NO and NO_2 concentrations during the day and from day to day inside the SRM office. Before the application of DtoxGuard® Int (14 January 2019 – 4 March 2019), the NO indoor concentration ranged from 0 to 125 ppb and was most of the time below 80 ppb. Several peaks above 100 ppb were measured and the NO indoor concentration reached exceptionally a peak at 177 ppb. Its mean value was 38 ppb. The NO_2 concentrations were lower than those for NO, between 2 ppb and 17 ppb with a peak at 21 ppb, and inferior to the outdoor limit defined for long-term exposure (40 μg/m^3 $\approx$ 21.3 ppb). Its mean value was 7.30 ppb. The evolution trends for both gases were similar to the ones reported in the literature. In an earlier study, Ekberg compared the supply air and the exhaust air in office buildings located in traffic-influenced environments. He reported NO_2 concentrations in the exhaust air for one building investigated: the values were lower than in the supply air and ranged from 6 to 24 ppb. For NO, the concentrations varied between 0 and 100 ppb, with a peak value at 180 ppb[10]. Challoner and Gill investigated the indoor air in commercial buildings located along busy street canyons in Dublin's city centre. They measured a daily variation in NO_2 concentration, which ranged from 5 to 30 ppb depending on the ventilation systems and NO_2 outdoor concentration[12]. Moreover, Mandin *et al*. sampled the air in office buildings across

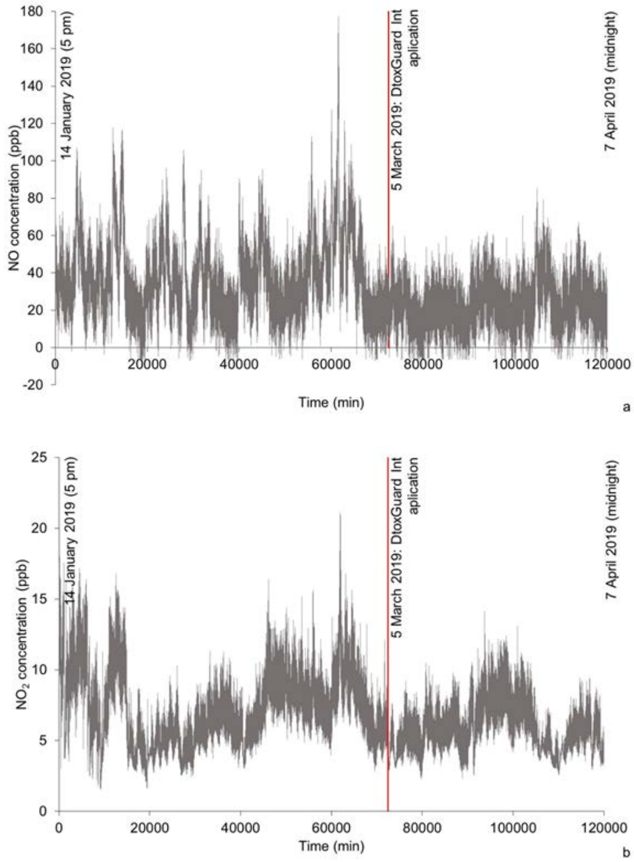

Figure 5. (a) NO indoor concentrations; (b) NO_2 indoor concentrations. The DtoxGuard® Int coating was applied to the wall on the 5^{th} of March 2019. Notice the different scales

Europe in summer and winter in the frame of the OFFICAIR project. For each building investigated they used a passive sampling technique, which did not allow measuring the daily variations as shown in Figure 5(a) and Figure 5(b). During the winter campaign, the minimum, maximum and mean NO_2 indoor concentrations were 4.8 μg/m^3 ($\approx$ 2.55 ppb), 39 μg/m^3 ($\approx$ 20.75 ppb) and 18 μg/m^3 ($\approx$ 9.57 ppb) respectively[28].

A change in the concentration evolutions occurred after the application of DtoxGuard® Int (6 March 2019 – 7 April 2019). For NO pollutant, a narrower range of variations was observed and only few peaks were monitored. The concentrations were mostly inferior to 50 ppb. For NO_2, although the change was less pronounced, the concentrations seemed to move towards lower values, mainly below 10 ppb. The mean values for NO and NO_2 were 23.43 ppb and 5.87 ppb, which corresponded to a decrease of roughly 38% and 20% compared to the period before the coating application (14 January 2019 – 4 March 2019).

3.2 NO and NO_2 outdoor concentrations

NO and NO_2 outdoor concentration evolutions collected by the two automatic air monitoring stations are shown in Figure 6(a), 6(b) respectively. These data were downloaded from the Air Quality England website for the period investigated, from 14 January 2019 to 7 April 2019[29]. The same pattern of evolution was observed for the two sites but the concentrations monitored by the station located at Oxford Road were much higher. Such amplitude differences can notably be explained by the short distance between the stations and the A57 Manchester road (Figure 2). Indeed, the influence of traffic-related air pollutants is expected to decrease with increasing distance of the site to the highway[30].

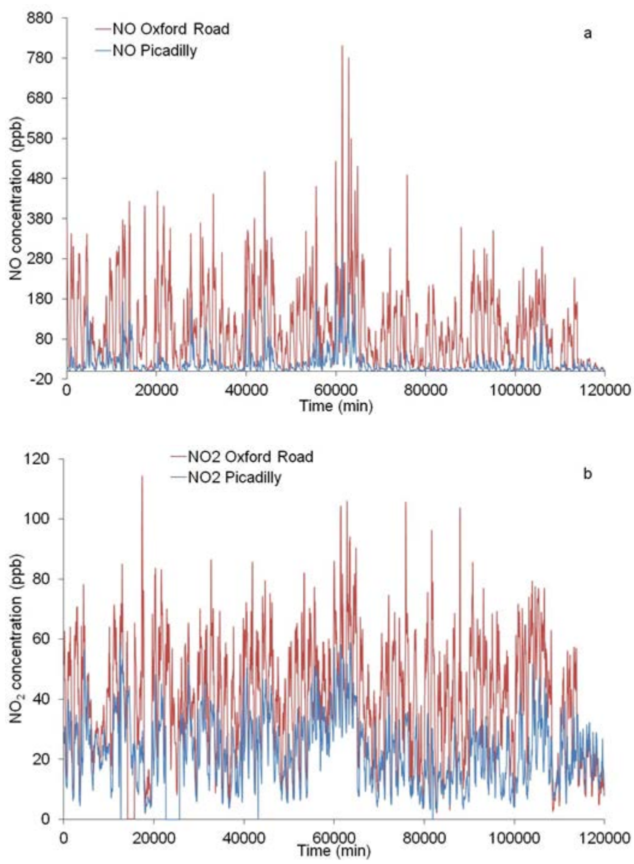

Figure 6. (a) NO outdoor concentrations monitored by the Oxford Road and Picadilly stations; (b) NO_2 outdoor concentrations monitored by the Oxford Road and Picadilly stations (data were downloaded from the Air Quality England website). Notice the different scales

The indoor and outdoor concentrations of NO and NO_2 are plotted in Figure 7(a), 7(b) respectively. The curves "Average NO outdoor" and "Average NO_2 outdoor" correspond to the means of the concentrations monitored by each station, as the SRM building is approximately halfway located. An indoor/outdoor correlation was observed as reported by previous studies[12,31–33]. Outdoor and indoor concentrations followed the same pattern, with indoor concentrations being lower.

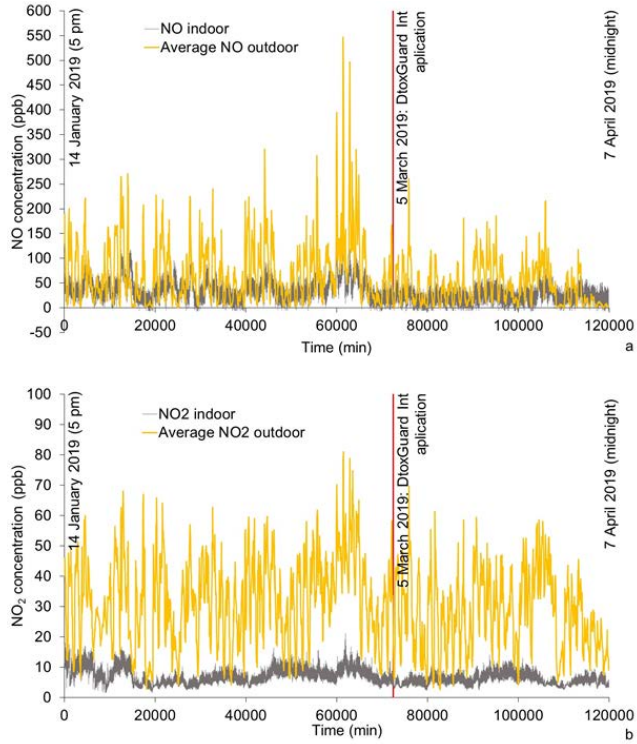

Figure 7. (a) NO outdoor and indoor concentrations; (b) NO_2 outdoor and indoor concentrations. The outdoor concentration curves correspond to the mean of the concentrations monitored by each station (cf. Figure 6). Notice the different scales.

Table 2 summarizes the mean and maximum values of indoor and outdoor concentrations for both gases and each period (before and after DtoxGuard® Int application). The indoor/outdoor ratios calculated from the mean values were also reported: $\approx$ 0.53 and $\approx$ 0.22 for NO and NO_2 respectively. With no contribution from indoor sources (as it could be the case in houses), outdoor concentrations, which were attenuated by the building envelope, were the primary factor in determining the indoor level of NO_x. Colbeck found that the indoor/outdoor ratios for NO_2 varied from 0.34 to 0.54 with an average value of 0.44 in two shops in Colchester (UK)[31]. The same trends were noted by Kukadia *et al.* who reported a reduced level of external pollutants such as NO and NO_2 inside two buildings in Birmingham (UK): the transient peak concentrations were approximately divided by two[32]. Moreover, Phillips et al. characterized three offices in Central London with respect to air quality. They found that outdoor levels of NO_x, NO_2 and NO exceeded indoor levels: the indoor/outdoor ratios calculated from the mean values were between 0.21 and 0.59 for NO_2 and 0.33 and 0.80 for NO[33].

Table 2. Mean and maximum values of indoor and outdoor concentrations for NO and NO_2 before and after DtoxGuard® Int application. The I/O (Indoor/Outdoor) ratios were calculated from the mean values

Values		Before DtoxGuard® Int application 14 January 2019 - 4 March 2019	After DtoxGuard® Int application 6 March 2019 - 7 April 2019
Mean values (ppb)	Indoor NO	37.98	23.43
	Outdoor NO	72.3	43.45
	Indoor NO_2	7.3	5.87
	Outdoor NO_2	32.51	27.62
I/O ratios (calculated from mean values)	NO	0.53	0.54
	NO_2	0.22	0.21
Max values (ppb)	Indoor NO	177.32	85.19
	Outdoor NO	547.34	260.75
	Indoor NO_2	21.1	14.15
	Outdoor NO_2	81.11	69.75

4 Discussion

4.1 Likelihood analysis

A decrease in NO indoor concentrations and, to a lesser extent in NO_2 indoor concentrations, was observed from the 6^{th} of March 2019 (Figure 5(a), 5(b) and Table 2). It could be due to: (i) the decrease in the outdoor pollution as both indoor and outdoor concentrations were correlated (cf. part 3.2), and/or (ii) the photocatalytic activity of the DtoxGuard® Int coating. In order to define the role of each phenomenon in changing the levels of NO and NO_2, and draw conclusions on the efficiency of the photocatalytic coating, the authors conducted further analyses of the raw data. They suggested comparing the likelihood of exceeding a certain concentration threshold before and after the DtoxGuard® Int application. Results are presented in Figure 8. The chosen thresholds were 40, 35, 20, 15 and 5 ppb for NO, and 15, 10, 5, 7.5 and 2.5 ppb for NO_2. The probability that the NO or NO_2 concentration was above the defined threshold during the period investigated was quantified by a percentage. Firstly, it can be noted that, for both gases, the indoor and outdoor percentages calculated before the DtoxGuard® Int application (from 14 January 2019 to 4 March 2019) reached higher values than the ones obtained after the DtoxGuard® Int application (from 6 March 2019 to 7 April 2019). The difference was less pronounced for the lowest thresholds (15 and 5 ppb for NO; 5 and 2.5 ppb for NO_2).

Moreover, the ratios between indoor and outdoor percentages were calculated for the two periods investigated (before and after the DtoxGuard® Int application). These values are summarized in Table 3 and presented on two radar charts in Figure 9. If the decrease in the outdoor concentrations was the only phenomenon at the origin of the observed decrease in the indoor concentrations after the DtoxGuard® Int application, similar indoor/outdoor ratios before and after the DtoxGuard® Int application should be expected for a same threshold. For NO, this was true for 5 ppb, 15 ppb and 20 ppb: the outdoor/indoor ratios were 1.143, 1.192 and 1.083 after the DtoxGuard® Int application and 1.064, 1.138 and 1.148 before the DtoxGuard® Int application. However, the ratios significantly decreased after the DtoxGuard® Int application for the 35-ppb and 40-ppb thresholds (0.249 and 0.136 respectively). They were 3 and 4.60 times lower than the ones before the DtoxGuard® Int application. In the case of NO_2, the ratios for the 7.5-ppb and 10-ppb thresholds were 2.60 and 15 times lower respectively. The differences between indoor/outdoor ratios after the DtoxGuard® Int application and those before the coating application are clearly visible on Figure 9 for the highest thresholds. This means that something else was happening, which led to a decrease in the peaks of the NO and NO_2 indoor concentrations and limited the concentration variations after the DtoxGuard® Int application. The authors reasonably thought that this was due to the photocatalytic activity. DtoxGuard® Int contains TiO_2 semiconductor particles, which were activated under the natural and artificial light in the SRM office. Redox reactions were then promoted to decompose NO_x. Such process has been well documented in the literature[24,34–36]. DtoxGuard® Int coating was also used in a previous study, in which it was applied to the walls (a surface of 9.3 m^2 was covered) in a 10-m^3 experimental chamber. The authors reported high NO degradation rates under UV and visible light[37,38].

Table 3. Ratios between indoor (I) and outdoor (O) percentages for each threshold

	Before DtoxGuard® Int application 14 January 2019 - 4 March 2019					After DtoxGuard® Int application 6 March 2019 - 7 April 2019				
Threshold (ppb)	40	35	20	15	5	40	35	20	15	5
I/O ratio for NO	0.626	0.742	1.148	1.138	1.064	0.136	0.249	1.083	1.192	1.143
Threshold (ppb)	15	10	7.5	5	2.5	15	10	7.5	5	2.5
I/O ratio for NO_2	0.004	0.166	0.442	0.809	0.996	0	0.011	0.17	0.682	1

4.2 Correlation coefficients

During this experimental campaign, other parameters than NO and NO_2 indoor concentrations were monitored by the AQMesh® station: O_3 concentration, temperature, humidity and atmospheric pressure. In order to assess the influence of these parameters on NO and NO_2 concentration variations, correlation coefficients (between two parameters) were calculated according to Equation 1. Table 4 summarizes the correlation coefficients (absolute value) obtained.

$$Correlation\ (X,\ Y) = \frac{\sum (x-\bar{x})\,(y-\bar{y})}{\sqrt{\sum (x-\bar{x})^2 \sum (y-\bar{y})^2}} \quad (1)$$

Where x and y are the parameters investigated and $\bar{x}$ and $\bar{y}$ are the mean values.

As shown in Table 4, the correlation coefficients between the investigated parameters were inferior to 0.5 except for NO_2 and O_3 concentrations, for which the coefficient was 0.717. The chemical coupling of these two gases has been discussed in previous studies[39–41]. Ozone is indeed a secondary photochemical pollutant that is produced from anthropogenic precursors, such as traffic emissions. NO_2 is mainly produced via NO in combustion processes by oxidation of nitrogen. NO_2 then acts as a precursor of O_3 and has a major impact on its daily variation: a reduction in the level of NO_2 is accompanied by an increase in the level of O_3. Such variation was notably discussed in[41]. To a lesser extent, the coefficients revealed a correlation between NO and NO_2 (0.368), and between both ga$_s$es with temperature (0.412 for NO/Temp. and 0.0337 for NO_2/Temp.)[42].

According to the authors, the correlation of NO and NO_2 with the other parameters could not explain the decrease in NO and NO_2 concentrations observed from the 6^{th} of March 2019. This analysis corroborated the previous one (cf. part 4.1), which attributed the decrease in the concentrations of NO and NO_2 to both above mentioned phenomena, *i.e.* the photocatalytic activity of the coating and the overall decrease in ambient air pollution.

Table 4. Correlation coefficients (absolute value). Each coefficient was calculated between two parameters. The parameters investigated were: NO, NO_2 and O_3 concentrations, temperature, humidity and atmospheric pressure. The highest correlation coefficient is in bold

	Time	NO	NO_2	O_3	Temp.	Humidity	Atm. Press.
Time	1	0.304	0.237	0.193	0.155	0.084	0.204
NO		1	0.367	0.105	0.412	0.03	0.264
NO_2			1	**0.717**	0.337	0.232	0.218
O_3				1	0.101	0.194	0.079
Temp.					1	0.056	0.247
Humidity						1	0.137
Atm. Press.							1

5 Conclusion

This case study provides detailed information on the IAQ in an open space office in Manchester, UK. The indoor concentrations of NO and NO_2 were monitored during 84 days, from 14 January 2019 to 7 April 2019 with the AQMesh® air quality station. On the 5^{th} of March, a photocatalytic coating was applied to a small part of an indoor wall. The collected data showed that the indoor and outdoor concentrations of NO and NO_2 significantly varied during the day and from day to day. The indoor concentrations were correlated to the outdoor pollution: the indoor/outdoor ratios were found to be around 0.53 for NO and 0.22 for NO_2. A decrease in NO and NO_2 concentrations was observed from the 6^{th} of March. The authors showed that this decrease was both due to the decrease in outdoor pollution and to the photocatalytic degradation of NO and NO_2. They notably assessed the likelihood that the concentrations of NO and NO_2 exceeded a certain threshold and compare the results obtained before and after the photocatalytic coating application. This analysis showed that the DtoxGuard® Int coating limited the concentration variations and reduced the peaks of NO and NO_2: the likelihood of exceeding 30 ppb for NO and 7.5 ppb for NO_2 was 3 times and 2.60 times lower after the coating application respectively. Given the influence on numerous parameters on NO_x concentration, this paper reported an original approach to assess the response of a photocatalytic coating in a real

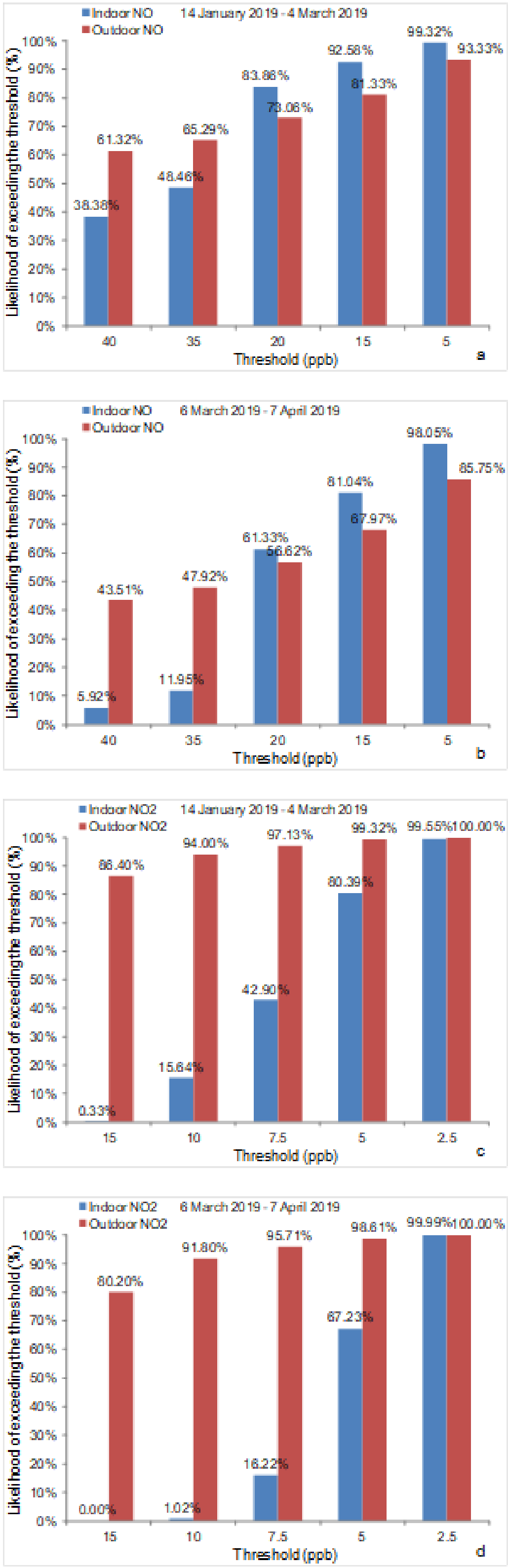

Figure 8. Likelihood of exceeding a concentration threshold: (a) for NO indoor and outdoor concentrations before the DtoxGuard® Int application; (b) for NO indoor and outdoor concentrations after the DtoxGuard® Int application; (c) for NO_2indoor and outdoor concentrations before the DtoxGuard® Int application; (d) for NO_2 indoor and outdoor concentrations after the DtoxGuard® Int application

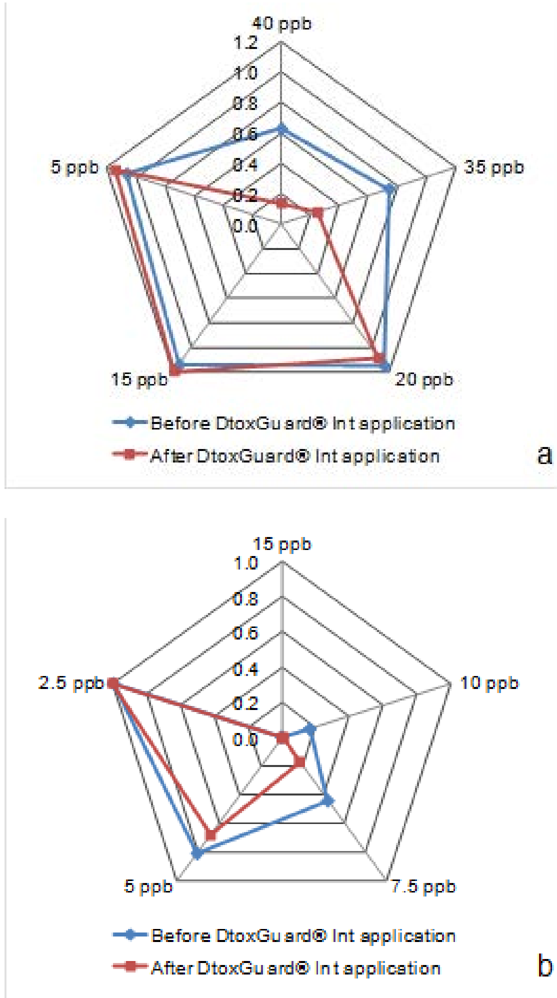

Figure 9. Radar charts showing the indoor/outdoor ratios obtained for each threshold before and after the DtoxGuard® Int application: a) for NO and b) for NO_2

indoor environment. It has to be kept in mind that the coated surface in the presented study was small (2.88 m^2), which means that a higher decrease in indoor pollution could be expected.

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Conflict of interests

The authors declare that there is no conflict of interest regarding the publication of this paper.

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