EFFECTS OF PHOTOCATALYTIC PAVEMENTS ON AIR QUALITY: REAL SCALE APPLICATIONS

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ABSTRACT

The latest pavement technologies and products have been developed with attention to their effects on the environment and health. Researchers have been studying solutions that could be termed "environmentally friendly". Nowadays many photocatalytic solutions have been proposed to counter air pollution, for example: cement, self-locking blocks, tiles, bitumen membranes and special products for asphalt mixtures. These solutions have always been laboratory-tested, in specific conditions and standards. They have rarely been analysed in real scale locations. Environmental conditions are very important for the photocatalysis process: this is the reason why the aim of this study was quantifying the reduction of some pollutants as nitrogen oxide (NO_x) in situ. Photocatalytic suspensions with titanium dioxide (TiO_2) were sprayed on road pavement at room temperature; two pavements were tested: the arrivals area in Milano Malpensa Airport (18,000 square meters) and a branch of Milano urban network (20,000 square meters). In order to verify the effectiveness of the treatments, the air quality of these areas was monitored for eight weeks. Pollutants concentration, temperature, solar radiation, relative humidity, wind speed and direction were registered continuously using normal monitoring equipments. Results obtained from comparing the data before and after spraying the photocatalyst emulsion demonstrated the effectiveness of the treatments, showing a significant reduction in the concentration of nitrogen oxide.

Keywords: photocatalysis, titanium dioxide, asphalt pavement, air quality, nitrogen oxide.

INTRODUCTION

Nowadays the phenomena of urban heat and pollutants are mainly due to traffic and can be considered a civil engineering matter. In particular NO_x (a group of highly reactive gases), nitric oxide (NO) and nitrogen dioxide (NO_2) and others pollutants originating from high-temperature fuel combustion EPA (2010) are responsible for many human diseases Kim et al. (2004) and a worsening urban heat phenomena. Even with recent advancements, ACEA (2011), the majority of vehicles are not making use of new engine technologies. As a result, health officials are moving for faster and more sustainable solutions to reduce the impact of traffic emissions. Increasing traffic volume and air pollutants concentrations have received significant attention during the last decades, leading to the need for a solution to purify indoor and outdoor air. Despite the emission control requirements and the installation of emission reduction systems, exhaust gas pollution by nitrogen oxides (NO_x) is a serious issue, especially in urban areas. A similar condition coexists in airport areas, where the level of emissions can easily exceed safe maximum concentrations. Even though the number of aircraft is vastly inferior to that of vehicles on the road, their effect is invariably worse. Furthermore, traffic congestion in the area of arrivals and departures can be assumed to be equal to that of a congested urban network.

One promising solution is to utilize pavement to withhold pollutants from the environment by means of an effective photocatalytic layer such as titanium dioxide (TiO_2) Fujishima et al. (2008). Existing applications presently include water purification, air conditioning (air purification), self-cleaning glazing, ceramic tiles (self-cleaning, antibacterial…), textile (anti-odour), mirrors (anti-condensation), tunnel lightning, white tents. Beldeens (2006). Therefore, considerable interest in innovative air pollution reduction systems was developed and some important advances have been made. The first applications were developed in Europe but the technology rapidly spread across to the United States, China and Canada. Most photocatalytic pavements use a photocatalyst (typically TiO_2) which is applied on the surface. Depending on the type of pavement, titanium dioxide may be present in the mix of cement concrete (for rigid pavements), sprayed or painted on the surface (for flexible pavements). In this sense, photocatalytic sprayed pavements are widely used in Italy. This technology adopts a chemical system to support the titanium dioxide and allow it to attach to the road surface.

In this paper, the development of the research on photocatalytic road materials is described, focusing on a real scale application of an area of pavement sprayed with a photocatalytic emulsion. It provides promising results in terms of a reduction in air pollutants concentration, measured by a test set-up (developed for the quantitative assessment of the air-purifying ability of photocatalytic road materials) and results from a test area.

SCOPE

The aim of this research was to evaluate the effectiveness of a particular photocatalytic emulsion on real scale applications. For this purpose, an area of road nearby the International Airport of Milano Malpensa (MXP) and a urban area in Milan (Viale F. Restelli-Via M. Gioia) were chosen to monitor the air quality and the concentration of pollutants emitted by traffic, before and after the application of the photocatalyst on the asphalt pavement.

PHOTOCATALYTIC EMULSION

Photocalytic emulsions are polymeric solutions of water plus a photoreactor (typically titanium dioxide in its anatase form Marwa et al. (2010)) in which the first is of a liquid form and the second is dispersed into it. Photocatalytic compounds such as nano-sized Titanium Dioxide (TiO_2) particles trap and absorb organic and inorganic particles in the air, removing harmful pollutants such as nitrogen oxides (NO_x) in the presence of UV light (sunlight). In addition, chemical additives can be added to the emulsion to improve the stability of the solution (improving the workability and regulating the breaking of the emulsion) and to avoid the contact between TiO_2 and the organic particles in the bitumen.

EXPERIMENTAL PROGRAM

Pilot Project

The research program focused on the validation of laboratory analysis through the application of the photocatalytic emulsion on two different, real traffic areas. In both cases the study lasted for eight weeks. The first four weeks were used as a reference. During this phase, the air quality was appraised on the effect of traffic only so the pollutant concentrations were set as the initial condition. In the others four weeks, with the application of the photocatalytic emulsion on the pavement, the effectiveness of the treatment was monitored.

The equipment used to measure the air quality included two mobile laboratories placed on two small road vans. Each mobile station was independent from the other and it was supplied with instruments to monitor the environmental conditions and the concentration of pollutants. The acquisition of data included environmental parameters such as: room temperature, direction and intensity of wind, relative humidity, irradiance and pressure. In addition, both stations were able to monitor the concentration of the main pollutants: nitrogen oxide (NO_x), carbon dioxide (CO_2), Ozone (O_3) and sulphur dioxide (SO_2). The acquisition of data was continuous (24h) and periodical checks were carried out from the data system in order to verify the status of equipment during the period of analysis.

Principally, the results demonstrated a reduction in the concentration of pollutants as a consequence of the application of the photocatalytic emulsion on the tested pavements. In order to have the afore-mentioned reduction it was important to consider the particular characteristics of the considered areas; it is especially important to remember that one of the areas was at an airport while the other one was in an urban environment.

TEST AREAS

Identification

The International Airport of Milano Malpensa (MXP) is located not far from the urban area of Milan city and is the second biggest airport in Italy; the main one in the North. From its two terminals, over 19 million passengers and thousands of aircrafts depart and land every year ASSAEROPORTI (2012). The tested area is about 18.000 m^2; it is situated between Terminal 1 and the Sheraton Hotel and it is covered by a glass roof (Figure 1a). This area is the most congested of the airport, in fact: it allows access to the terminal from the highway, entry to the main parking garage and the Sheraton Hotel nearby. Most of the paved surfaces are in asphalt concrete and they are used not only as car transit and parking but also by pedestrians, resulting in heavy traffic congestion.

The second area studied (Viale F. Restelli) is situated in Milano not far from the Central Railway Station and the Lombardy Regional Government buildings; this area it is about 20.000 m^2 (Figure 1b).

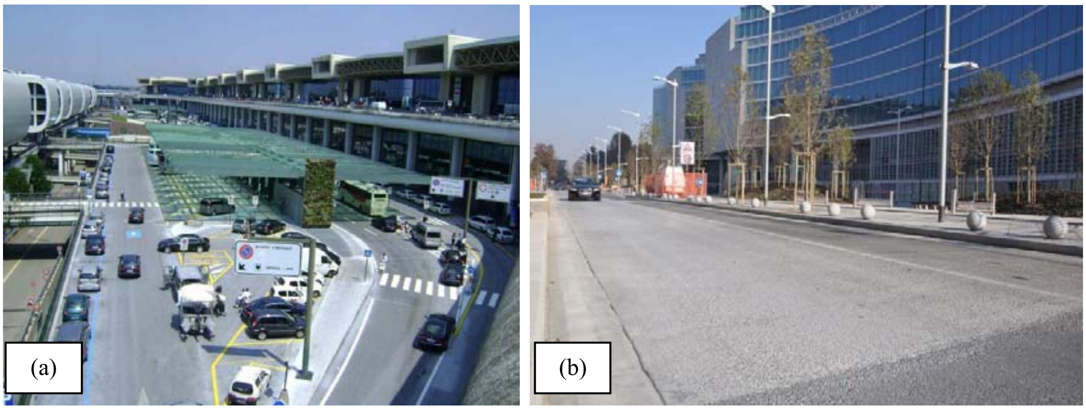

Figure 1: The treated areas: (a) Arrival area of MXP Airport (b) Viale F. Restelli in Milan's urban center

Application of the photocatalytic emulsion

Since the traffic of the Malpensa area is largely congested and chaotic, the laying of the photocatalytic emulsion was carried out during the night. Specifically, the area was sprayed over two nights of May 2011 while the photocatalytic emulsion in Viale F. Restelli was applied in the middle of October. In both cases the photocatalytic emulsion was applied with a spray-bar mounted on the rear of an urban van, without any heating (at room temperature). The dosage was monitored through the forward speed and the number of open nozzles on the bar (Figure 2).

Figure 2: The application of the photocatalytic emulsion: a) Arrival area of MXP Airport (b) Viale F. Restelli in Milan's urban centre

Before spraying the photocatalytic emulsion, the surfaces were cleaned with a traditional road sweeper and most of the particles and dust were removed. Once spread, the emulsion started to break, allowing the molecules of water to separate from those of TiO_2 and increasing the adhesion property of the paved surface. The curing time required to re-open the areas to traffic ranges from half an hour to several hours, depending on the temperature and relative humidity. In the case of the Malpensa area, warm temperatures and low humidity-reduced the curing to only one hour while in the case of Viale F. Restelli it was necessary to repeat the application after an unexpected rainfall so the curing took longer.

Installation of mobile stations

To ensure the best results in terms of data reliability, two mobile stations were installed in both cases: one mobile station (Station 1) was placed in the middle of the treated areas, specifically in the Arrivals area of Malpensa airport and in Viale F. Restelli in Milano, while the second (Station 2) was parked far from the experimental field (in the middle of the Sheraton Hotel area for the airport case and in Via M. Gioia for the urban one). During the first four weeks, the stations monitored the environmental condition of the untreated areas. After the application of the photocatalytic emulsion (week 5), Station 1 started to register the changes impacting on the air quality.

Sensitivity Analysis

The first phase of the research included a sensitivity analysis to ensure the photocatalytic effectiveness of the treatment. A series of core samples were taken from pavements and brought to the laboratory. All specimens were tested in accordance with the Italian Standard UNI 11247, so the reduction index could be computed. The test setup was built and calibrated as described by Crispino et al. (2011). The pollution reduction was quantified in reference to NO, NO_2, and NO_x loss of concentrations in ppb (part per billion) from the reaction chamber. Results of the analysis showed that the presence of photocatalytic emulsions produced a significant reduction in NO_x concentration in all tested samples (Figure 3a, b).

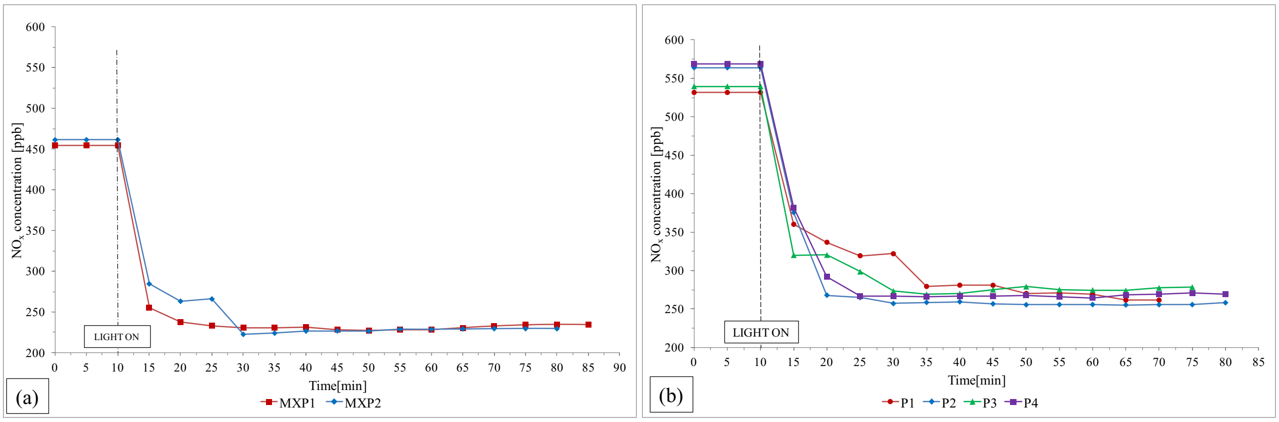

Figure 3: Laboratory test results: NO_x concentration reduction over time: (a) MXP (b) Milan

RESULTS AND ANALYSIS

Table 1 shows the concentration of pollutants and the operating conditions of test areas employed to measure the NO_x degradation over time. Two types of parameter are listened: (i) room variables and (ii) concentration of non reactive pollutants. For the room variables, solar irradiance (I), and relative humidity (RU) were considered since they have a fundamental role in photocatalytic processes. As for the pollutants, it was decided to report only non-reactive substances in order to distinguish them from the pollutants involved in the process of photocatalysis (such as NO and NO_2). The concentration (C) of sulphur dioxide (SO_2), carbon monoxide (CO) and ozone (O_3) are reported separately for the mobile stations located in the reference zones (Sheraton Hotel and Via M. Gioia) and the treated zones (Arrivals and Viale F. Restelli).

Table 1: Results of air quality testing from Malpensa International Airport and Milan city. Room temperature (T), relative humidity (RU), solar irradiance (I), wind speed (V_{wind}), height of rain (h_{rain}), concentration of pollutant (C_i).

Week No.	T [°C]	RU [%]	I [W m^{-2}]	V_{wind} [m s^{-1}]	h_{rain} [mm]	C_{SO2} [µg m^{-3}]*		C_{CO} [mg m^{-3}]**		C_{O3} [µg m^{-3}]*	
	MALPENSA AIRPORT					SHERATON	ARRIVALS	SHERATON	ARRIVALS	SHERATON	ARRIVALS
1st	15.0	39.7	225.6	1.6	0.3	1.0	3.9	1.0	0.6	71.9	53.3
2nd	17.4	56.8	222.3	1.3	1.6	4.6	4.5	1.0	0.9	77.3	66.8
3rd	16.4	63.5	203.7	1.5	9.0	3.2	4.3	1.0	0.9	67.3	54.6
4th	18.4	47.7	270.2	1.3	0.1	4.3	5.1	1.0	0.9	87.6	68.2
5th	20.2	47.0	255.6	1.6	13.0	4.2	5.8	0.9	0.9	82.5	60.7
6th	23.7	45.6	264.9	1.4	0.2	4.6	5.4	0.9	1.2	96.1	66.2
7th	20.3	57.3	209.4	1.8	34.2	3.9	4.6	0.9	1.0	79.4	54.3
8th	19.3	74.8	168.8	1.4	108.8	3.0	3.2	1.0	0.9	54.8	0.9
	MILAN					GIOIA	RESTELLI	GIOIA	RESTELLI	GIOIA	RESTELLI
1st	23.3	54.1	97.8	1.0	0.0	5.6	4.7	1.4	1.6	38.8	42.7
2nd	18.8	40.7	88.7	2.0	0.0	7.3	8.0	1.2	1.2	30.1	28.9
3rd	14.8	53.0	93.2	1.5	0.9	5.8	6.4	1.1	1.2	24.9	25.4
4th	11.7	66.2	75.8	1.3	33.5	4.3	4.3	1.2	1.2	15.2	18.0
5th	13.9	72.7	84.1	0.9	0.5	6.0	5.6	1.7	1.4	8.9	19.0
6th	14.6	80.1	24.5	1.9	125.0	4.0	5.1	1.1	1.2	22.6	23.2
7th	9.6	75.9	44.7	1.1	0.3	6.0	5.1	1.8	1.9	8.6	9.8
8th	7.4	77.7	28.9	1.0	0.7	9.0	9.4	2.4	2.6	4.1	7.0

*(microgram per cubic meter) **(milligram per cubic meter)

Figure 4 shows the variation in NO and NO_2 maximum concentrations from the test sites. Both pollutants showed a sensible variation over time, but no single trend. In the case of NO_2, the curve seemed to be insensible to the measuring station. On the other hand, NO showed a significant dependence to the presence of the photocatalytic treatment (from week 5). Since the weather conditions at the mobile stations were the same, the NO concentration was a direct consequence of the thermic combustion of vehicles, whilst NO_2 reacts with the first. The photocatalytic oxidation mechanism of NO_x is reported in several publications Ballari et al. (2011). All of them propose that NO oxidation to NO_2, and then NO_2 to HNO_3 is due to the attack of the hydroxyl radical generated in the photocatalyst activation stage. It is well known that photocatalytic oxidation can be divided into three main steps: mass transport and absorption of pollutants from the air mass to the surface of the catalyst; photocatalytic reaction to the catalyst; and desorption and mass transport of the reaction products from the surface of catalyst to air Chen et al. (2009). In particular the purifying rate of NO is obviously higher than NO_2 Chen and Chu (2011), and this can be explained with reference to the numerical results reported in Figure 4. It is an explanation as to why the concentration of NO was so variable.

In addition, the relationships between the average hourly concentrations of air pollutants (as NO_2, O_3, SO_2, CO, and PM10) has been clearly demonstrated by Kim et al. (2004), so numerical values from NO and NO_2 were further influenced by the number of vehicles that transited the road sections during the period of analysis.

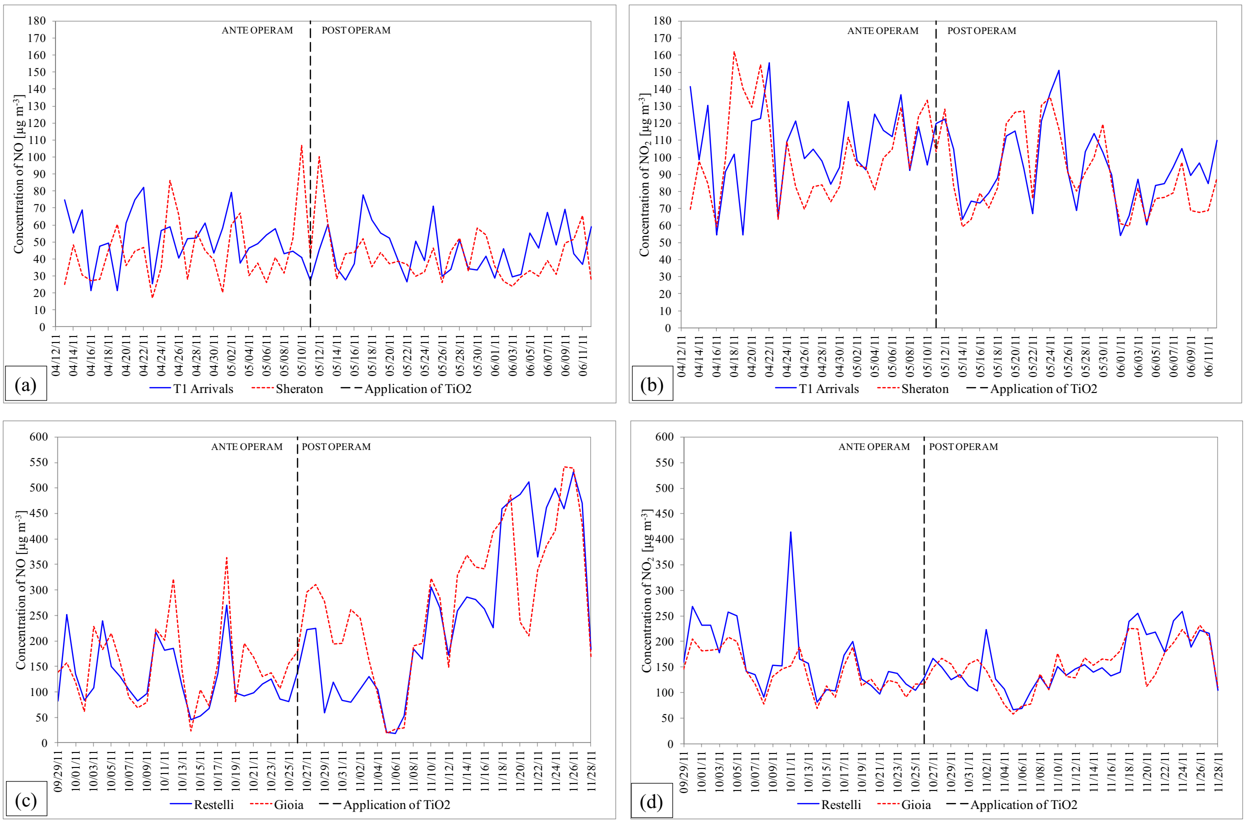

Figure 4: Maximum daily concentration of NO and NO_2: a), b) Malpensa Airport c), d) Milan.

Due to this the analysis of the concentration of pollutants before, and the application of the photocatalytic emulsion after (ante and post operam) produced inconsistent results. In fact, the comparison can be easily influenced by exogenous factors such as differing weather conditions or atmospheric emissions due to variation in traffic flow. As such, data may not be comparable, and comparison could lead to errors of overestimation or underestimation due to the weather.

Therefore, it was necessary to find a parameter to eliminate the disparity of boundary conditions. In this sense, the concentration of pollutants has been expressed as the average ratio of concentrations measured by mobile stations over time:

$$A(t) = \frac{c_{pollutant,location\ 1}}{c_{pollutant,location\ 2}} \qquad (1)$$

where C is the concentration of specific pollutant and "location" is the measuring point. In order to obtain comparable curves, location 1 always refers to the treated area (Arrivals or Viale F. Restelli) while location 2 to the untreated or reference sections (Sheraton Hotel or Via M. Gioia). Under this hypothesis, the weather conditions are identical for both locations, the coefficients in Eq. 1 are affected equally and concentrations can be easily compared.

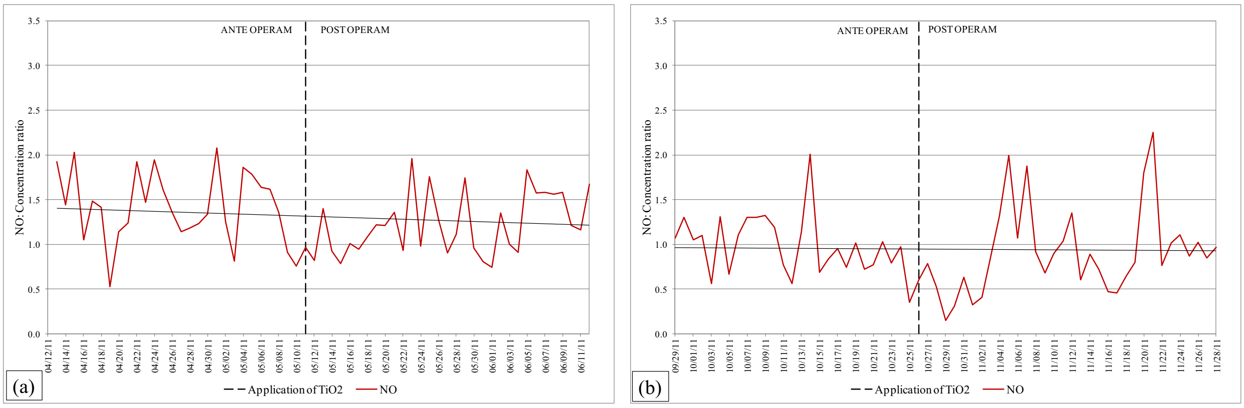

Figure 5: Evolution of NO ratio: (a) Malpensa (b) Milan

The results from the calculation of NO ratio are shown in Figure 5. By representing the pollutant ratio via a regression line, it is evident how NO decreases over time. In this sense, the slope of the line shows the decrease in NO while the boundary values of the segment show the initial and final conditions. Through this hypothesis one can see that the reduction in NO was around 15% for Malpensa (Figure 5a) and 5.5% for Milan (Figure 5b).

Figure 6 shows the evolution of the NO_2 ratio. Decreases in ratio over time are indicative of a variation of the boundary conditions. In particular the emissions and the type of vehicles (road vehicles or aircrafts) strongly influenced the curve of NO_2. In addition, it is well known that the chemical transformation of NO to NO_2 is faster than the transformation of NO_2 to HNO_3 so the purifying rate of NO is higher than NO_2, and this can be explained by reference to NO_x complete reaction Chen and Chu (2011). In fact, as demonstrated by Ballari et al. (2011), when both contaminants NO and NO_2 have similar concentrations, the NO oxidation reaction is slightly faster than the NO_2 degradation. However, when the relative humidity is high, then the photocatalytic reaction rates are slower and NO_2 is produced in the system, most likely because the photocatalytic oxidation of NO, producing NO_2 is moderate Ballari et al. (2011). In line with this, results from Malpensa (Figure 5a, 6a) indicate a period with high values of irradiance and low humidity, as demonstrated by Table 1.

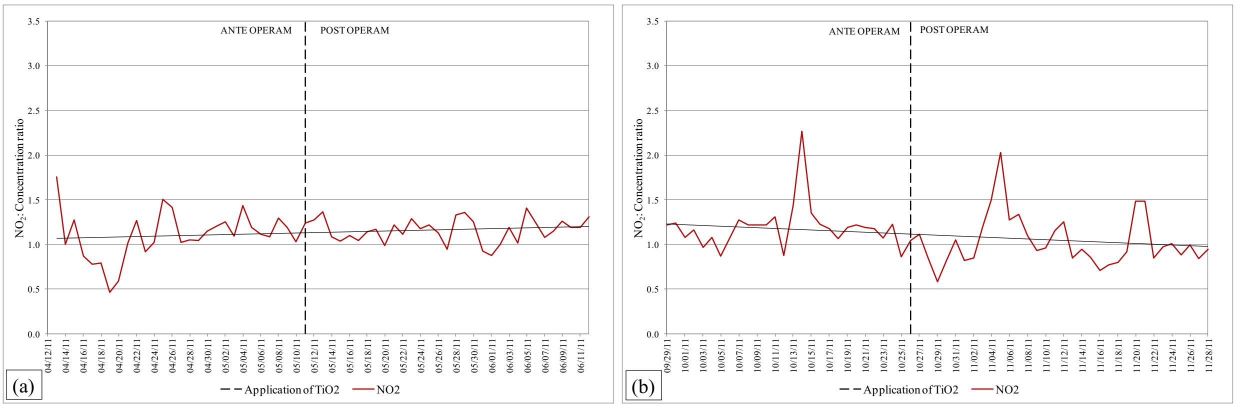

Figure 6 Evolution of NO_2 ratio: (a) Malpensa (b) Milan

Since in a standard atmosphere, the reactions of NO and NO_2 are usually considered as a group, the results of the NO_x ratio as the sum of NO and NO_2 ($mg.m^{-3}$) are reported in Figure 7. Note, that from the observation of these graphs, the NO_x ratio was quite constant (+0.7%) for Malpensa while it decreased by 8% for Milan, hence no significant conclusion can be easily drawn. Detecting and quantifying the contribution made by the traffic to local concentrations of NO_x is extremely difficult, in particular referring to a period of only eight weeks.

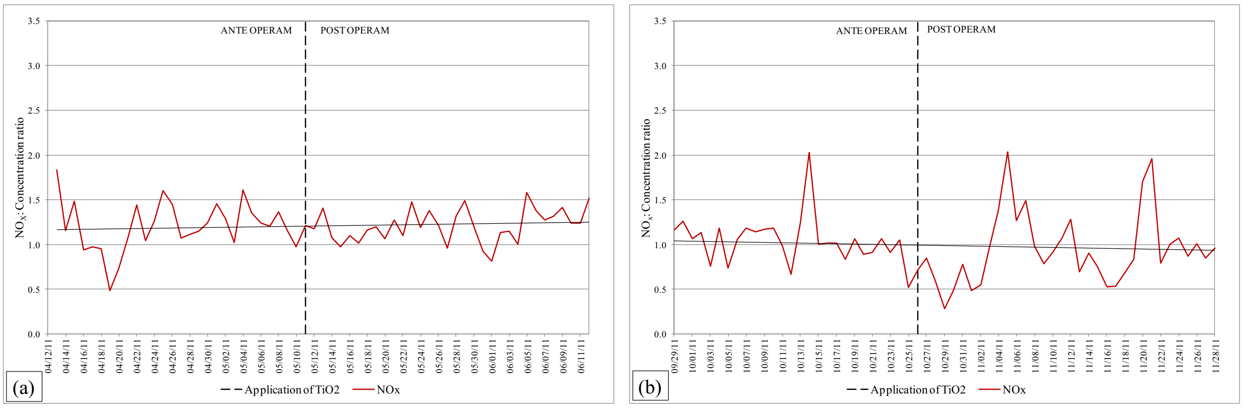

Figure 7: Evolution of NO_x ratio: (a) Malpensa (b) Milan

Nevertheless, even if the ratio is unaffected by the weather changes, it is still influenced by the traffic. In particular it is well known that NO_x are produced by traffic and its concentration depends on the number and type of vehicles and motion condition. Since the traffic data were not available it was necessary to identify a suitable variable to characterize the traffic flow. In addition, the variable must be independent from the weather in order to register the changes in emissions of nitrogen monoxide during the two periods (ante and post operam).

In this sense the variation of traffic conditions was quantified with reference to a reduction in the SO_2 and CO ratios since they are non reactive pollutants and they are not influenced by the weather. Sulphur dioxide is found to be a good marker of aircraft emissions Carslaw et al. (2006), due to its relationship to aircrafts traffic flows. In fact, it is a non-reactive pollutant with a significant impacts upon human health. It is not strictly influenced by weather conditions, and its concentration at the two mobile stations can be representative of traffic on a micro scale.

On the other hand, Carbon monoxide poisoning is the most common type of fatal air poisoning in many countries. It is a temporary atmospheric pollutant in some urban areas, chiefly from the exhaust of internal combustion engines. Yu et al. (2004) showed that CO and its concentration can be related directly to road traffic, and therefore to traffic flows in urban centers. Owing to this, it was assumed that SO_2 and CO can be considered representative of traffic flows.

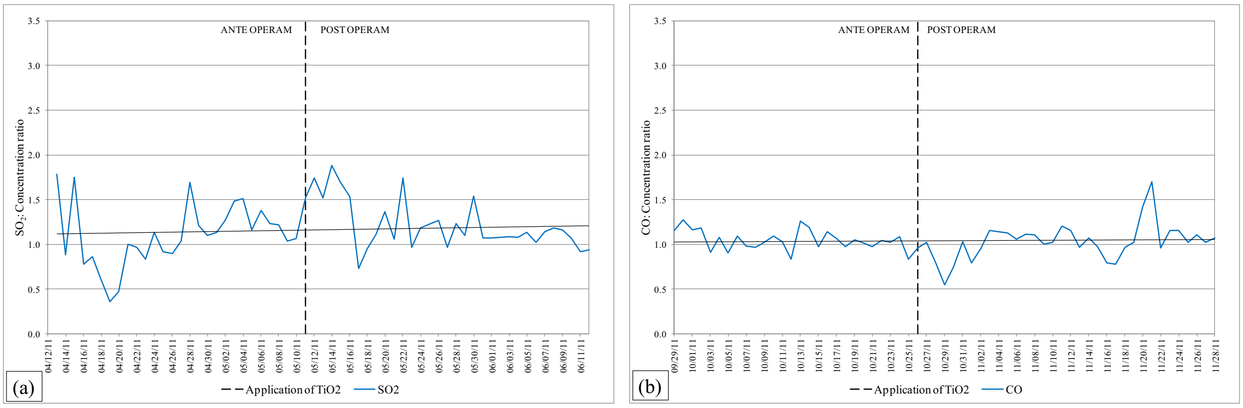

Figure 8: Evolution of the SO_2 and CO ratios

As shown in Figure 8a-b, the increment in SO_2 was approximately 15% as opposed to the 0.95% increase in CO. Given this, the increase in sulphur concentration corresponds to an increase in traffic flow at Malpensa, while the constant rate of carbon monoxide shows that the traffic has not changed in Milan during the course of the study. To determine the effective reduction in pollutant concentration within the two sites, it was necessary to normalize all data to the traffic flow. As such, the gradient of the all curves presented in Figures 5 and 6 has been adjusted to the referring pollutant: SO_2 for Malpensa Airport and CO for Milan. From the application of the normalization procedure, it was possible to compare results. The results of the measurement of pollutant concentrations during the real scale applications are shown in Table 2.

Table 2: normalized reduction of pollutants

Normalized concentration ratio	NO	NO_2	NO_x
Malpensa (T1 Arrivals – Sheraton Hotel)	25%	9%	9%
Milan (Viale F. Rerstelli – Via M. Gioia)	5%	7%	12%

The results obtained for the area of Malpensa showed a significant decrease in NO in contrast to the small reduction in NO_2 with a total NO_x reduction of 9%, whereas in Milan the reductions of NO and NO_2 were inverse but the amount of total NO_x dropped by 12%. This was probably because under different irradiances the oxidation of NO to NO_2 changes in line with different percentages of humidity and vehicles emissions.

CONCLUSION

In the present work, NO and NO_2 photocatalytic reactions are reported through the investigation of two areas. In the first case, a road section nearby Terminal 1 of Malpensa International Airport was treated with a photocatalytic emulsion. In the second case, the same emulsion was laid down on a urban road in the centre of the city of Milan. Detecting and quantifying the contribution made by the traffic to local concentrations of NO_x is difficult, therefore the ratio of the average concentrations was used with the aim of eliminating boundary conditions. From the obtained results, the following conclusions can be drawn:

- The impact of the reductions in NO_x concentration owing to the photocatalytic emulsion was quantified through a sensitive laboratory analysis. The tests conducted in accordance with the Italian Standard UNI 11247 showed a significant reduction in nitrogen oxide levels.
- The real scale applications confirmed that this photocatalytic emulsion is an effective technology for air purification as proven by the registered concentration of pollutants during a measuring period of eight weeks.
- A simple evaluation of air quality data cannot guarantee satisfying results since pollutant concentration can be affected by the environmental conditions. In this sense deepen analysis must be carried out to eliminate the influence of traffic and weather on the measurements taken at the mobile stations.
- The adoption of the concentration ratio as a reference parameter allowed the elimination of variations due to weather, since both terms of the ratio were affected equally.
- SO_2 and CO are non-reactive pollutants, whose concentrations are not affected by weather conditions. The differences in their concentrations are thus exclusively due to differences in traffic volume on a micro scale. As a result of this, it was possible to normalize the reduction of nitrogen monoxide considering the effective variation of traffic volume during the period of measurement.
- With reference to results from Malpensa and Milan, the reduction in pollutants was significant. In fact the purifying rate of nitrogen ranged from 5% to 25% depending on the pollutant and the site in question.
- The presented outcome demonstrates the effectiveness of the photocatalytic emulsion on both test areas. Therefore further analysis is required in order to confirm results over an extended period of measurement and different environmental conditions..

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