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Effects of TiO_2 based photocatalytic paint on concentrations and emissions of pollutants and on animal performance in a swine weaning unit

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ABSTRACT

The beneficial effect of a TiO_2-based photocatalytic treatment on the indoor air purification of a swine farm has been evaluated in a trial performed in two identical mechanically ventilated traditional weaning units, with 391 animals lodged in each of them. One unit was used as reference, whereas the walls of the second unit (260 m^2) were coated with *ca.* 70 g m^{-2} of TiO_2 and irradiated with ten UV-A lamps. The environmental parameters (*i.e.* the ventilation rate, the internal and external temperature and relative humidity), together with NH_3, CH_4, CO_2 and N_2O concentrations in the exhaust ducts and PM_{10} emissions, were monitored in the two units throughout all of the production cycle (75 days). Significant decreases in CH_4 concentration (*ca.* 27%, $P < 0.05$) and PM_{10} emission (*ca.* 17%, $P < 0.01$) were observed, together with an increase of the piglets' productive performance in the treated unit with respect to the reference one. Indeed, the ADG (Average Daily Gain of piglets) was 424 g *vs.* 414 g for the piglets lodged in the two units, respectively, with a significantly better feed conversion ratio (FCR, ratio between the food ingested by the animals and their weight gain) of 2.18 *vs.* 2.44 ($P < 0.001$). Therefore, the photocatalytic treatment with TiO_2 coating had positive effects not only on methane concentration and particulate matter concentration and emission, but also significantly improved the feed conversion ratio of growing piglets, very likely due to the increased quality of indoor air, with positive economic repercussions for the farmer. Internal photocatalytic treatment in swine husbandry could thus be considered as a potential Best Available Technology (BAT).

1. Introduction

Large scale animal husbandry is a great source of environmental air pollutants, such as ammonia, greenhouse gases and dust. A lot of air quality studies have shown that the gases released from swine manure are likely to lower the quality of indoor air with consequent decrease in swine performance and environmental air quality (Osada et al., 1998).

Photocatalysis with semiconductors has been successfully employed as a tool to treat polluted water and air (Hoffmann et al., 1995; Fujishima et al., 1999). Photocatalytic reactions on semiconductors are initiated by the absorption of light, so that highly reactive species are formed on their surface, *i.e.* the electrons photopromoted in the semiconductor conduction band and the so-called holes in the valence band. TiO_2 is the semiconductor by far most widely employed in photocatalytic applications, thanks to its outstanding stability and high activity. Valence band holes produced in TiO_2 under irradiation have a high oxidation potential, which has been largely exploited in oxidation processes (Lee and Choi, 2002; Lee et al., 2002). TiO_2 photocatalytic treatment may be applied not only to organic substrates, but also to inorganic species, in either oxidation and reduction reactions (*e.g.* precious metals recovery from water solutions). The most widespread industrial research on TiO_2 photocatalysis is aimed at using its oxidation properties, for potential uses in sterilisation, sanitation, and anti-pollution applications in the aqueous phase (Murgia et al., 2005) and in the atmosphere (Levine and Calvert, 1977). In particular, TiO_2 photocatalytic paints can be used to transform NH_3 into N_2, N_2O or NO and water (Il'Chenko and Golodets, 1975). Particularly in the case of ammonia present in toxic industrial wastewaters, for which biological treatments are not appropriate, photocatalytic oxidation, leading to ammonia removal as molecular nitrogen, may be envisaged as a promising alternative purification technology.

However, only a few scientific papers appeared so far on the use of TiO_2-based photocatalytic paints and coatings for pollutants and

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noxious gases abatement (Allen et al., 2005), particularly for zoo-technical applications. Only recently, the effects of TiO_2 photocatalysis were studied in livestock houses, with extremely encouraging results (Guarino et al., 2008).

Aim of the present study is assessing the effects of photocatalytic TiO_2-painted walls on the air quality inside a swine weaning unit, on both the emissions of particulate matter (PM_{10} and $PM_{2.5}$) and NH_3 and greenhouse gases release into the environment, in relation to the parallel effects on the animal productive performance.

2. Materials and methods

2.1. Location

The trial was performed in two identical mechanical ventilated weaning units in a swine farm in Northern Italy. 391 animals were lodged in each unit with an initial mean weight of 8.9 kg (8.3–9.5 kg). Pigs left the weaning unit at the mean weight of 41.66 kg (41.43 kg–43.89 kg), 48 days after their lodging as weaners. One unit was used as a control unit, while the other unit was treated with TiO_2 to test the photocatalytic effect.

2.2. TiO_2 coating procedure and lamps installation

A transparent and odourless TiO_2 liquid suspension was sprinkled on the inside walls of the treated unit. The liquid solution, Activa – PPS® (Proactive Photocatalytic System Technology) is produced by Global Engineering S.p.A. In the Safety Data Sheet (S5-300B, October 13 2003) available from the producer it is reported that this product has no negative effects on animal and operator health. A coating amount of 70 g m^{-2} was used, over a total internal surface of 260 m^2. Ten solar spectrum lamps (G13, Poker by Plexiform, Italy; 122 cm long) were installed in the test unit, in two rows 2.25 m above the floor, transversely positioned as reported in Fig. 1. They simulate sunlight, emitting UV-A radiation, in the 315–400 nm wavelength range with a power consumption of 36 W per lamp. They were always kept on during the experiment. The green isocurves in Fig. 1 indicate the light intensity values measured at the ceiling level.

2.3. Environmental parameters and animal performance measurements

The environmental parameters (*i.e.* temperature and relative humidity), the ventilation rate, the gas and particulate matter concentration were monitored in the two units throughout all the production cycle together with the animal productive performance in terms of growth rate (Average Daily Gain, ADG) and feed conversion ratio (FCR, ratio between the food ingested by piglets and their weight gain).

The building was mechanically ventilated. The incoming air was provided by two perforated pipes (40 cm diameter placed longitudinally in the unit (see Fig. 2)). Fresh air came from the corridor of the

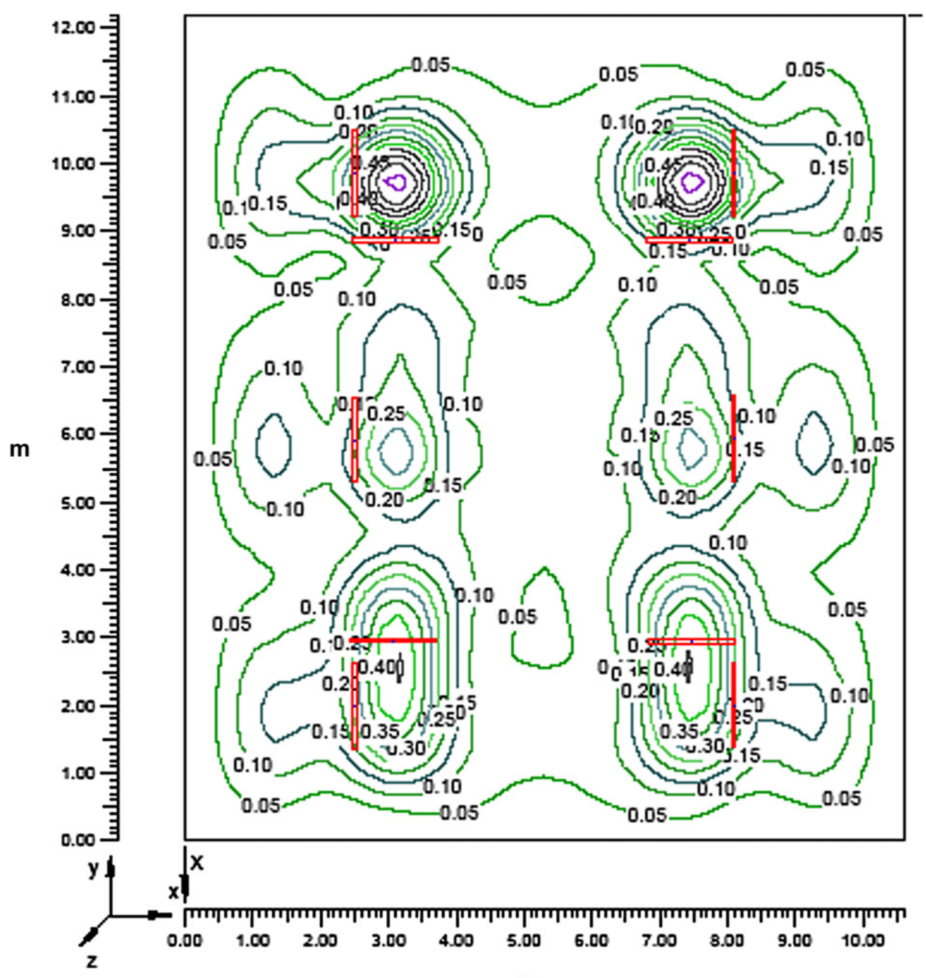

Fig. 1. Distribution of the light intensity (lx) measured above the lamps in the experimental unit coated with TiO_2.

Fig. 2. One of the two units of the trial. The two perforated pipes (p) placed longitudinally in the unit and linked to the corridor of the building provide fresh air; the exhaust air is removed by two fans (f).

building, the exhaust air was removed by two fans (10,000 $m^3 h^{-1}$ maximum ventilation rate). The chosen set-point of the internal temperature in the farm units is automatically controlled by the Fancom FMS type controller by using the ventilation rate throughout the compartments.

2.4. Ammonia, greenhouse gases and particulate matter concentration

The concentration of NH_3, CH_4, CO_2 and N_2O was measured *in continuo* in the exhaust ducts of both units, using an infrared photoacustic detector IPD (Brüel & Kjaer, Multi-gas Monitor Type 1302, Multipoint Sampler and Doser Type 1303) collecting data every 15 min, measurements were performed as described by Costa and Guarino (2009).

The concentration of PM_{10} and $PM_{2.5}$ was continuously monitored by two identical instruments (HAZ DUST-EPAM 5000), which combine the traditional gravimetric technique with the so-called "near-forward light scattering". The light scattering of an infrared radiation is employed to immediately and continuously measure the concentration of airborne dust particles, in mg/m^3. An infrared light source is positioned at a 90° angle from a photo detector. As the airborne particles enter the infrared beam, they scatter light and the amount of light received by the photo detector is directly proportional to the aerosol concentration. The signal is internally processed and compensated for noise and drift. This allows high resolution, low detection limits and excellent base line stability.

The inlet system of the instruments was configured to sample particulate matter and the PM_{10} or $PM_{2.5}$ impactors were placed, each on one of the two dust samplers.

The pollutants emission factor is usually estimated as the product between the daily mean concentrations of bio-aerosols and the daily mean ventilation rate, in $m^3 h^{-1}$ (Berckmans et al., 1991). By this way, the emission of aerial contaminants emitted from pigs' buildings have been calculated in previous literature (Seedorf and Hartung, 2001, 1998; Takai et al., 1998). The emission factors are expressed per Livestock Unit (LU), equivalent to 500 kg of body weight.

The emission rate was calculated as the product between the pollutant's concentration and the ventilation rate recorded at the same time, according to the following equation:

Equation 1. Pollutant's emission calculation:

$$E_i = C_i \times V_i \quad (1)$$

where: E_i = Pollutant's Emission at time i, C_i = Pollutant's Concentration at time i, V_i = Ventilation rate at time i.

The error of the pollutant emission factor (δE) is limited by the sum of the errors of the pollutant's concentration measurement (δC) and the ventilation rate measurement (δV).

Equation 2. Error of the pollutant's emission factor:

$$\delta E = \delta C + \delta V \quad (2)$$

2.5. Statistical analysis

Variance analysis (ANOVA) was performed on the collected data using SAS statistical package (SAS 9.1, 2009) to investigate the effects of TiO_2 coating on indoor pollutant concentrations and emissions from the piggery and on animal performance, keeping also as effects ventilation rate and environmental parameters recorded in the two units.

3. Results and discussion

3.1. Piglets' performance

The productive performance was higher in TiO_2-coated and illuminated unit, where ADG was 424 g *vs*. 414 g in the reference unit; the FCR was 2.18 *vs*. 2.44 ($P < 0.001$; see Fig. 3).

3.2. Environmental parameters

The temperature and relative humidity did not show any significant difference in the two units. The mean temperature value recorded in the reference unit was 25.9 °C (temperature ranging from 24.4 °C up to 29.7 °C) and the mean relative humidity was 63% (58 %–96 %). In the test unit the mean temperature was 25.7 °C (recorded values ranging from 24.2 °C to 29.9 °C) and the relative humidity was 56% (52–90%).

The mean ventilation rate was 15,870 $m^3 h^{-1}$ for the reference unit and 16,263 $m^3 h^{-1}$ for the TiO_2-coated unit. The ventilation rate variation during the trial is shown in Fig. 4.

3.3. Concentration of ammonia, greenhouse gases and particulate matter

The mean values of ammonia and nitrous oxide concentration were similar in the two units (around 1.9 mg m^{-3} and 1.48 mg m^{-3},

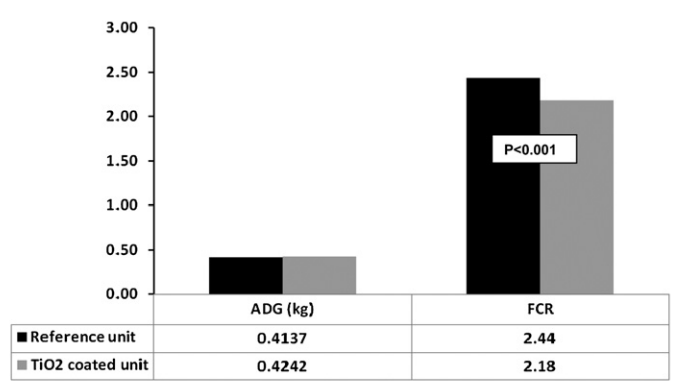

Fig. 3. Productive performance of piglets in the two units.

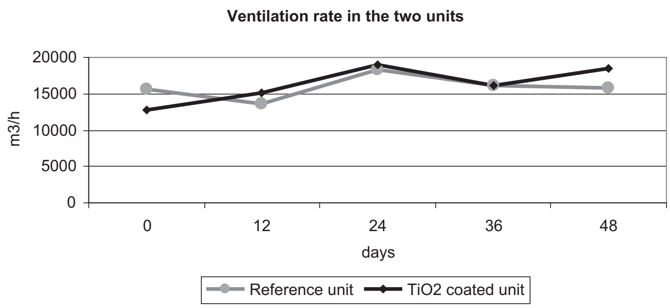

Fig. 4. Ventilation rate during the trial in the two units.

Table 2
Average emission of pollutants from the two units.

Pollutant	Reference unit ($g\ h^{-1}$)	Experimental unit ($g\ h^{-1}$)
NH_3	30.267	29.755
N_2O	23.341	23.518
CO_2	32991	34463
CH_4	261.718[a]	190.068[b]
PM_{10}	0.914[A]	0.770[B]
$PM_{2.5}$	0.236	0.255

(a, b) Values in the same row followed by different letters are significantly different for $P < 0.05$. (A, B) Values in the same row followed by different letters are significantly different for $P < 0.01$.

respectively, see Table 1). Carbon dioxide mean values differed, even if not in a significant way, with a lower concentration in the reference unit. Methane showed a significantly higher concentration in the reference unit (16.63 mg m^{-3} *vs.* 12.04 mg m^{-3}, $P < 0.05$) and the same occurred for PM_{10} (0.058 mg m^{-3} *vs.* 0.048 mg m^{-3}, $P < 0.01$). The fraction of particulate matter lower than 2.5 μm, *i.e.* $PM_{2.5}$, did not exhibit any difference in the two units (concentration was around 0.02 mg m^{-3} in both units).

Ammonia concentration was higher in both units during the first period of observation (2.06 mg m^{-3} for the reference unit *vs.* 2.77 mg m^{-3} for the TiO_2-coated unit) until the second week, while in the following days of the trial it declined reaching the values of 1.44 mg m^{-3} *vs.* 1.23 mg m^{-3} respectively.

The same trend was shown by nitrous oxide (1.19 mg m^{-3} for the reference unit *vs.* 1.76 mg m^{-3} for the TiO_2-coated unit) and carbon dioxide (1734 mg m^{-3} for the reference unit *vs.* 2312 mg m^{-3} for the TiO_2-coated unit), with higher concentrations in the initial phase of the trial and lower values in the middle and final parts of the experimental study: at the last sampling day mean nitrous oxide concentration was 1.84 mg m^{-3} for the reference unit *vs.* 1.48 mg m^{-3} for the TiO_2-coated unit, mean carbon dioxide concentration was 2416 mg m^{-3} *vs.* 2455 mg m^{-3} for the photocatalytic treated room.

Methane concentration was similar in the two units up to the second week (16.32 mg m^{-3} for the reference unit *vs.* 15.93 mg m^{-3} for the TiO_2-coated unit), but in the following days remarkably lower mean values were detected in the TiO_2-coated unit (17.32 mg m^{-3} *vs.* 9.98 mg m^{-3}). The concentration of methane increased during the trial in the reference unit, as expected, due to the slurry permanence in the pit.

PM_{10}, *i.e.* particles less than 10 μm in diameter, posing a health concern because they can be inhaled into and accumulate in the respiratory system, on average showed a higher concentration in the TiO_2-coated unit in the first part of the experimental trial.

However, starting from the third week, their concentration was lower in the TiO_2-coated unit, following a trend almost similar to those of the other pollutants considered in the experiment.

3.4. Ammonia, greenhouse gases and particulate matter emission

Statistical analysis performed on pollutant concentrations and emissions, keeping into account values of ventilation rate, which, being higher in the experimental unit (see Fig. 4) could induce a dilution of pollutants concentration, confirmed the positive effects of the photocatalytic treatment on air quality in the treated unit.

As reported in Table 2 and Table 3, significantly higher values of methane and PM_{10} emissions were found in the untreated reference unit, under similar environmental conditions and with an identical number of piglets per unit. The reduction of methane emission was estimated around 27%, with mean values of 190.07 g h^{-1} for the treated unit *vs.* 261.72 g h^{-1} ($P < 0.05$) for the reference unit.

Also the emission of particulate matter, in terms of PM_{10}, was 16% lower as a result of the photocatalytic treatment, with mean values of 0.770 g h^{-1} for the treated unit *vs.* 0.914 g h^{-1} for the reference unit. The values for gases and dust are referred to emissions originated by 391 piglets with a 23 kg mean weight, corresponding to 18 LU (LU, Livestock unit = 500 kg).

In a previous study (Guarino et al., 2008), higher effects of TiO_2 coating and photocatalytic treatment on the environmental pollutants were observed in a farrowing unit, on both ammonia concentration and emission. This could be due to the different type of manure removal in the farrowing unit tested in that study (*vacuum system*), while, in the present case, manure was collected in a deep pit under the slatted PVC floor.

Indeed, the two units employed in the present study were located in a traditional swine intensive husbandry with a manure storage system under the floor. This represents a "traditional" structural solution, usually characterized by higher concentration and emission of gases, above all of methane, in comparison with modern solutions based on "BAT" (Best Available Technology) manure system removal.

In fact, as reported by IPCC Emission Factor Database (Costa, 2010a,b), the methane emission factor, calculated on yearly basis,

Table 1
Average concentration of pollutant in the two units.

Pollutant	Reference unit ($mg\ m^{-3}$)	Experimental unit ($mg\ m^{-3}$)
NH_3	1.90	1.92
N_2O	1.48	1.48
CO_2	2075	2134
CH_4	16.63[a]	12.04[b]
PM_{10}	0.058[A]	0.048[B]
$PM_{2.5}$	0.02	0.02

(a, b) Values in the same row followed by different letters are significantly different for $P < 0.05$. (A, B) Values in the same row followed by different letters are significantly different for $P < 0.01$.

Table 3
Average emission of pollutants from the two units (in LU units).

Pollutant	Reference unit ($g\ d^{-1}\ LU^{-1}$)	Experimental unit ($g\ d^{-1}\ LU^{-1}$)
NH_3	40.36	39.67
N_2O	31.12	31.36
CO_2	43988	45951
CH_4	348.95[a]	253.42[b]
PM_{10}	1.218[A]	1.026[B]
$PM_{2.5}$	0.315	0.34

(a, b) Values in the same row followed by different letters are significantly different for $P < 0.05$. (A, B) Values in the same row followed by different letters are significantly different for $P < 0.01$.

is 24.57 g d^{-1} LU^{-1} in a weaning unit with vacuum system manure removal. Emission values of 348 g d^{-1} LU^{-1} and 253. 42 g d^{-1} LU^{-1} were recorded, respectively, for the reference unit and the treated unit of the present study. Such an unusually high methane concentration mostly depends on the presence of the deep pit in the weaning units and on the unfrequently removed manure. In fact, while ammonia emissions are related to the emitting surface of manure (Ni et al., 1999), the methane production level is directly dependent on the time of manure permanence in the pit (Philippe et al., 2007), as confirmed by our experiments.

The positive effect of the photocatalytic painting in reducing methane concentration and emission evidenced in the present study is very important. In fact, although its photocatalytic oxidation product, *i.e.* carbon dioxide, is widely considered for its warming potential, its contribution to the greenhouse effects is less important than those of CH_4 and N_2O, whose warming potentials are, respectively, 23 and 296 times higher than that of CO_2 (Intergovernmental Panel on Climate Change, IPCC, 2005).

The concentration and emission rate of nitrous oxide were high in both rooms monitored in the trial, *i.e.* 23.34 g d^{-1} LU^{-1} for the reference unit and 23.52 for the experimental unit, respectively, whereas the emission of nitrous oxide measured on yearly basis in a weaning room with vacuum system for manure removal was reported to be around 3.62 g d^{-1} LU^{-1} (Costa, 2010a,b; Costa and Guarino, 2009). Also Phillips et al. (1995) reported that the N_2O emission rate from livestock units was 0.27 g h^{-1} LU^{-1}, or 6.48 g d^{-1} LU^{-1}, and that it could not be detected, being below measurement sensitivity, for rooms with partially slatted floors in summer.

The PM_{10} emission factors determined in the present study (1.026 g d^{-1} LU^{-1} for the treated unit and 1.218 g d^{-1} LU^{-1} for the reference unit) confirm that TiO_2 coating can remove PM_{10} in a livestock house, while a "traditional" BAT cannot potentially reduce particulate matter concentration and emission (Fabbri et al., 2007).

The mechanism involved in PM_{10} reduction is not yet clearly faced in literature, but several articles described the photocatalytic elimination, in air and/or in water, of polycyclic aromatic hydrocarbons (Zhang et al., 2008) dioxins (Choi et al., 2000), sulphate (Zhao et al., 2009), sulphur compounds (Cantau et al., 2007), bacteria, spores and viruses (Wu et al., 2009). All these products, which are among the constituents of PM_{10}, with carbon, mineral dust, pollen, nitrates (Seedorf et al., 1998), could compose the PM_{10} fraction lowered by the photocatalysis in this experiment, in agreement with a recent review by De Richter and Caillol (2011).

The environmentally critical conditions and the poor air quality of the system under study, with very high methane and particulate matter concentrations, clearly took advantage from the here investigated photocatalytic treatment, which thus seems to be most effective for high pollutant concentrations. In the present trial this occurs for methane, whose concentration is high because of the long-term presence of manure in the deep pit of the stable.

4. Conclusions

The results of the study provide evidence that the photocatalytic treatment, based on TiO_2 coating and UV-A light irradiation, has beneficial effects on methane and particulate matter concentration and emissions in the swine husbandry; the piglets feed conversion ratio was significantly improved by the photocatalytic treatment, most probably in relation to the better air quality of the treated unit.

The results obtained in the present, as well as in the previous study (Guarino et al., 2008), point to the conclusion that the indoor photocatalytic treatment of swine husbandries could be considered as a new potential low cost Best Available Technology.

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