Odour control versus potential pollution and risks

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Odours can cause hazards to the environment and human health. The increasing public odour complaints and expectations, and stricter environmental regulations have made odour control important all over the world. There are various technologies for odour control, mainly divided into physical, chemical, and biological treatment. For a sustainable solution of odour problems, there is more to consider besides the performance of the technologies to remove the target pollutants. This article focuses on the potential pollution and the environment-health risks that may be created during the odour control processes, including activated carbon adsorption, chemical scrubber, thermal treatment, biological treatment, UV photodegradation, and non-thermal plasma. Accordingly, the solutions to decrease the pollution and risks are discussed.

1. Odour control

Odours can be caused by a variety of harmful substances, such as H_2S , NH_3 , and volatile organic compounds (VOCs), which lead to a threat to the environment and human health. There are significantly increasing public complaints due to odorous emissions from municipal activities, such as waste water treatment plants, or industrial activities (Schlegelmilch et al., 2005b, Lebrero et al., 2011). As a result, the regulations related to air emissions have become stricter in many countries in recent years. For instance, in China, there was a project called the 12^{th} Five Year Plan (2011–2015), which clearly aimed to improve the quality of water and air (NPC, 2011).

A variety of technologies have been developed for odour control. According to the nature of each technology, they are mainly classified into physical, chemical, and biological technologies (Schlegelmilch et al., 2005b). To select an appropriate odour control technology, it is necessary to consider the nature and concentration of odorants, the level of efficiency required, the type of odour sources (active or passive), the air flow rates to be treated, site characteristics including operation and maintenance capabilities, etc. (Lebrero et al., 2011, Burgess et al., 2001). Moreover, there should be more to consider besides the factors mentioned above (especially not only the performance of the technologies to remove the target pollutants). The potential pollution and environment-health risks that may be induced during odour control processes should not be neglected. To the authors' knowledge, there are few works that focus on the potential pollution and the environment-health risks of each technology for odour control. In this study, we review the potential problems of pollution and risks of odour control processes, including activated carbon adsorption, chemical scrubber, thermal treatment, biological treatment, UV photodegradation, and nonthermal plasma. Moreover, the according solutions are discussed.

2. Potential problems and solutions

2.1 Activated carbon adsorption

2.1.1 Potential problems

Porous carbonaceous materials, such as activated carbon, are widely used in the removal of pollutants present in liquid and gaseous effluents due to their excellent adsorbent properties and large surface areas. They do not destroy the pollutants, but just concentrate them (Kittrell et al., 1991).

However, once exhausted, activated carbon can become hazardous residues. The Environmental Protection Agency (EPA, USA) defines that solid wastes generated during the treatment, storage, or disposal of a listed hazardous waste or their mixture with a listed hazardous waste are also hazardous waste. Therefore, activated carbon wastes from the treatment of contaminated air that contains listed hazardous wastes should be managed as hazardous waste. Spent activated carbon is usually incinerated or disposed in landfills, resulting in risks of pollution (Salvador et al., 2015a). The disposal of hazardous waste in landfills is less and less accepted due to the growing concerns about the effect of pollutants on the environment and more stringent environmental standards (Yuen and Hameed, 2009, Zanella et al., 2014). Adsorbates from granular activated carbon can be released into the atmosphere in reactive, oxidized and partially oxidized states, aggravating the air pollution (Nath and Bhakhar, 2011). The current Anniston Chemical Agent Disposal Facility operating permit states that used carbon will be evaluated for chemical agent contamination if it has been exposed to agent concentrations of $\geq 0.0001 \text{ mg/m}^3$. Once the carbon is determined to be a hazardous waste, it should be sent to a permitted hazardous waste treatment, storage, and disposal facility (NRC, 2009).

In addition to the disposal, the spent activated carbon can be regenerated, which attempts to remove pollutants retained on the activated carbon's surface to restore adsorptive capacity without modifying porosity or causing adsorbent mass losses. However, the removal process will result in either water or air streams polluted with the contaminants adsorbed by the activated carbon, and the liquid or air stream can be a disposal problem. The desorbed contaminants must be treated or reclaimed (Kittrell et al., 1991).

Regeneration methods include three major groups: thermal, chemical and microbiological regeneration (Salvador et al., 2015a, Salvador et al., 2015b). Thermal regeneration is the most widely used industrial-scale method (Salvador et al., 2015a). Thermal regeneration is typically processed at 800~1000 °C, which involves high energy consumption (Yuen and Hameed, 2009). Román et al. (2013) analysed the gas released during the thermal regeneration of spent activated carbon, and found that there were H_2 , CO, CH_4 and CO_2 in the gas. CH_4 and CO_2 are both greenhouse gases, which could enhance global warming (Sommerfeld et al., 1993), while CO is poisonous. Regeneration with liquid water implies that the fluid becomes contaminated by the same pollutants, subsequent purification of the liquid should be carried out (Salvador et al., 2015b).

Complete regeneration of the spent activated carbon using microbial processes is difficult to achieve, because microbial degradation products will be adsorbed and the pores will be blocked by the decay products of microbial cells (Nath and Bhakhar, 2011, Kim et al., 2014). Moreover, its ecological impacts should be further investigated.

2.1.2 Solutions

To make the saturated activated carbon re-usable and to avoid the contamination that is caused by its disposal, the regeneration of spent activated carbon is an attractive option, even though problems can arise during regeneration (Salvador et al., 2015a, Salvador et al., 2015b).

Regarding the high energy consumption of thermal regeneration, microwave energy can be a potential method and powerful replacement (Yuen and Hameed, 2009).

Adsorption can be used as an enrichment step prior to thermal gas treatment (Rafson, 1998), and it can be combined with thermal treatment or other treatments for the subsequent treatment of CH_4 and CO.

2.2 Chemical scrubber

2.2.1 Potential problems

Chemical scrubbers are the most commonly employed abatement techniques in waste water treatment plants (Gabriel and Deshusses, 2004). Chemical scrubbers are implemented in packed towers, using NaClO, NaOH, KMnO₄, H₂O₂, or H₂SO₄ as reagents. Considerable amounts of chemicals are required, and reaction products must be removed or treated. Chemicals used in chemical scrubbers for off-gas treatment might also generate residual odours as well as odorous reaction products such as aldehydes. The hazardous nature of the employed chemicals and the generated by-products represents a serious challenge to its supremacy in a world increasingly devoted to sustainable development (Schlegelmilch et al., 2005b, Charron et al., 2004, Gabriel and Deshusses, 2004, Jeavons et al., 2000, Metcalfeddy et al., 2003).

The use of chemical scrubbers can lead to climate change, freshwater eutrophication, photochemical oxidant formation, human toxicity and ecotoxicity, mainly due to the use of large amounts of chemicals. The use of chemicals in chemical scrubbers can account for approximately 80% of its total impact on climate change, with NaClO presenting the largest contribution. The chemicals added in chemical scrubbers (especially NaClO) can induce ecotoxicity and human toxicity. Moreover, hypochlorite has the tendency to oxidise and chlorinate the compounds at the same time. Furthermore, chlorine gas (Cl₂) is also generated (Schlegelmilch et al., 2005b). Even though NaCIO is both relatively inexpensive and easy to handle, the significant amount required for odorant oxidation can play a key role in the impacts associated with the use of chemicals (Alfonsín et al., 2015). In addition, surface water criteria to protect aquatic life (IDAPA 58.01.02.250) prescribes that pH should be within the range of 6.5 to 9.0 (EPA, 2016). However, the pH value of discharged water from the chemical scrubber is usually out of that range (Gabriel and Deshusses, 2004). Therefore, the use of chemicals in chemical scrubber could lead to negative burdens (Estrada et al., 2011, Alfonsín et al., 2013).

2.2.2 Solutions

As NaClO plays a key role in the impacts associated with the use of chemicals (Alfonsín et al., 2015), alternative chemicals, such as ferric and ferrous ions chelated with EDTA, Fe_2O_3 and FeO, and H_2O_2 , can be potential alternatives (Shareefdeen et al., 2005). The investigations on alternative chemicals, which are more environmentally friendly and cost-effective, could be conducted.

The chlorine gas formed in the chemical scrubber could be absorbed by NaOH in the same or another scrubber (Drust and Deacon, 1995).

The pH of the discharged water from the chemical scrubber should be regulated by adding acid or alkali (Drust and Deacon, 1995). If there are multiple stages, acidic waste water could be used to neutralize the alkali one, thus reducing the consumption of chemicals.

2.3 Thermal treatment

2.3.1 Potential problems

Thermal treatment can be basically applied to any exhaust air. Thermal treatment includes catalytic and non-catalytic techniques. In a non-catalytic thermal treatment process, VOC-laden air is thermally treated (oxidized/decomposed) at temperatures of about 730-850 °C (Chuah and Warahena, 2009). Since the concentration of VOCs is often low, it usually requires the addition of natural gas or a pre-concentration, such as by adsorption. However, this method produces secondary emissions like nitrous and sulphur oxides (Schlegelmilch et al., 2005b). In presence of NO_x , photochemical oxidation can happen (Alfonsín et al., 2013).

If the waste gas contains chlorine, the production of chlorine during the thermal treatment process is not desired, since it can create an air pollution problem. However, even the most highly active oxidation catalysts can oxidize the HCl product, and the product is chlorine rather than HCl (Kittrell et al., 1991).

Little attention has been paid to the production of dioxin-like compounds (DLCs), which have been classified by the World Health Organization (WHO) as one of the most persistent toxic chemical substances in the environment (WHO, 2002). The main risk of dioxins for human health is that they can alter the development of many cells and can cause illnesses, such as cancer, disruption of the endocrine system, or reproductive and development problems. The main sources of DLCs are the human interaction, especially combustion-related processes. PCDD/Fs are formed during the reactions between organic compounds and chlorine at high temperature (Dopico and Gómez, 2015). Several investigations have proven that DLCs can be generated during the thermal waste gas treatment (Li et al., 2012, Teng et al., 2015).

Therefore, some researchers indicate that incineration technologies are not an attractive odour abatement alternative due to their high investment and operation costs and the potential production of toxic dioxins and furans (Delhoménie and Heitz, 2005).

2.3.2 Solutions

To reduce the additional fuel, catalytic process can be an alternative, because they can be operated at lower temperatures (Zhang et al., 2016). Besides, for non-catalytic processes, using advanced systems with heat recovery (recuperative thermal oxidisers, regenerative thermal oxidisers) can reduce the energy consumption significantly (Greco and Jain, 2016).

 NO_x can be controlled by selective catalytic reduction (SCR), using NH_3 . V_2O_5/TiO_2 -based catalysts (Wielgosiński, 2011).

The selectivity of catalysts for oxidation of halogenated hydrocarbons is very important, because appropriate catalysts can convert the chlorine content to HCl rather than to Cl₂ or other chlorine-containing products (Kittrell et al., 1991).

The proper selection of raw materials to avoid the addition of chlorine into the process is an effective method to reduce the formation of dioxins. Extensive research and field measurements have indicated that commercial NO_x control catalysts also reduce the PCDD/F emission (Wielgosiński, 2011, Dopico and Gómez, 2015). Another way to

prevent the regeneration of DLCs is the application of the accelerated cooling of the off-gas (UNEP, 2006).

2.4 Biological treatment

2.4.1 Potential problems

Biological treatment is regarded as an environmentally friendly and cost-effective waste air treatment method. It simply uses microbes to consume the pollutants from contaminated air streams (EPA, 2003). However, biological treatment systems may also be a source of pollution.

Biological processes may produce high concentrations of bioaerosol, which are a potential health risk. Some investigations have been conducted on the sanitary consequences for workers. The results indicate an increase in respiratory, gastrointestinal, and skin symptoms compared with a control group (Schlegelmilch et al., 2005a). Ottengraf and Konings (1991) reported that there was an extra contamination due to the filtration process at low CFU-numbers of the inlet gas, therefore, biofilters may serve as a source of emission of microorganisms. Wang et al. (2009a) measured bioaerosol concentration of exhaust gas from the biofilter to investigate the ecological safety of the biological process. The result shows that the microorganism emission from the biofilter is as high as 1.38 10³ CFU/m³. Schlegelmilch et al. (2005a) found that the number of mesophilic bacteria determined in the outlet of the biofilter is about two times higher than that in the inlet, and microbial analysis revealed that the species of microorganisms at the inlet are different from that at the outlet of the treatment system. All these investigations have proven that biological treatment systems can cause secondary emissions of bioaerosol.

Besides the emissions of bioaerosol, biological treatment systems can also produce pollutants in the waste water. Sulphurous and nitrogenous organic or inorganic compounds are oxidized to sulphuric and nitric acid, which can cause acidification of water (Hartikainen and Ruuskanen, 1996). In addition, N-compounds discharged in the waste water, including free ammonia, ammonium, nitrite and nitrate, can result in water eutrophication (Alfonsín et al., 2013).

2.4.2 Solutions

Ozone is a biocide, which can control the bioaerosol emissions from the biological treatment system and the growth of excess biofilm in the bioreactor. UV photodegradation process produces ozone. Therefore, an integrated UV-biofiltration system is investigated, and proved to be a promising option to effectively reduce the bioaerosol emissions from biological treatment system (Wang et al., 2008, Wang et al., 2009b, Moussavi and Mohseni, 2007, Wang et al., 2009a).

The emitted concentration of bioaerosol is considerably affected by the structure of the packing material. Finely structured, homogeneous packing materials would emit lower CFU-numbers of bacteria than more inhomogeneous materials. Therefore, the selection of packing materials and optimization of its structure could be an approach to control the bioaerosol emissions (Ottengraf and Konings, 1991).

The rate of the bioaerosol emissions process is assumed to be proportional to the gas velocity. There is a clear decrease trend of the outlet concentration as the velocity increased. Thus bioaerosol emissions could also be reduced by controlling the gas velocity (Ottengraf and Konings, 1991).

Regarding the acidification and eutrophication caused by the formation of sulphuric acid, nitric acid and other N-compounds, the waste water from the biological treatment

system should be treated (neutralized or biological treated) before it is discharged into the aquatic environment.

2.5 Thermal treatment

2.5.1 Potential problems

UV photodegradation, using UV irradiation to rapidly oxidize the pollutants, is capable of oxidizing a wide range of contaminants (Koh et al., 2004).

A few studies indicate that some products, which are more toxic compared to their parent contaminants, can be generated during UV photodegradation (Koh and Thiemann, 2001, Cantavenera et al., 2007, Jeong et al., 2005, Zhang et al., 2006). Wang et al. (2009a) used UV photodegradation to treat chlorobenzene and found that both the acute toxicity and the genotoxicity of the photodegradation products increased in the exhaust gas after UV photodegradation. The exhaust gas containing these toxic products shows potential health risks. Cantavenera et al. (2007) indicated that genotoxic intermediates can be formed during the photocatalytic degradation of paraquat.

UV lamps (185- and 254-nm UV light) can produce a high concentration of ozone (up to 50 mg/m³). Moreover, it is reported that 0.2 mg/m³ of ozone might have a harmful effect on human health. What's more, the half-life of ozone in air is approximately 16 h (20 °C, 1 atm). As a result, health risks for workers are to be expected if the ozone processes self-decay in the gas phase (Moussavi and Mohseni, 2007, Wang et al., 2008, Wang et al., 2009a).

The possible production of toxic contaminants should be carefully taken into consideration when a UV photodegradation process is going to be applied.

2.5.2 Solutions

The toxic products, such as chlorophenol and ozone, produced by the UV photodegradation, can be removed by subsequent biofiltration. A biological treatment system shows an apparent detoxification effect on UV photodegradation. The combined UV-biofiltration process can be a promising option to make UV photodegradation ecologically safer (Wang et al., 2009a, Wang et al., 2008, Wang et al., 2009b). However, a biological system as an ozone deletion process should be used carefully, because ozone might inhibit or even kill microorganisms in the biological system (Moussavi and Mohseni, 2007).

2.6 Non-thermal plasma

2.6.1 Potential problems

Non-thermal plasma provides a highly chemical reactive environment (e⁻, O^{*}, HO₂^{*}, OH^{*}, N₂^{*}, O₃, etc.) to decompose the VOCs at ambient conditions (Sultana et al., 2015, Vandenbroucke et al., 2011).

Non-thermal plasma can result in the formation of excess ozone, which is a hazardous substance to human health (Pekárek, 2003). Moreover, the industrial implementation of non-thermal plasma for the abatement of VOCs is impaired by three main bottlenecks such as poor product selectivity, formation of undesired by-products (O_3 , NO_x , other VOCs and aerosols) that often increase the overall toxicity of the treated gas stream (Sultana et al., 2015).

2.6.2 Solutions

To decrease the undesired by-products, the combination of non-thermal plasma with catalysts/sorbents, i.e., plasma-catalysis, has been remarkably investigated during the

last decade (Sultana et al., 2015). In such a hybrid system, the catalyst can be integrated either inside (IPC-Inside Plasma Catalysis) (Kim et al., 2006) or downstream (PPC-Post Plasma Catalysis) (Vandenbroucke et al., 2011, Durme et al., 2008) of the discharge region. Mok and Kim (2011) tested a γ -Al₂O₃ packed dielectric barrier discharge reactor for the treatment of toluene in a sequential approach. The result shows that ozone is the only by-product besides CO and CO₂.

As far as ozone removal is concerned, the combination with a biological treatment system can be an option. Wei et al. (2013) combined non-thermal plasma and a biological process to treat dimethyl sulfide, and found that ozone from non-thermal plasma makes the microbial community in the biotrickling more active for dimethyl sulfide removal.

3. Conclusions

The increasing public odour complaints and expectations and stricter environmental regulations have motivated the development of odour control technologies. Besides the performance of removing target contaminants of each technology, the potential pollutions and risks should be noticed. This work aims to point out the potential pollution and environment-health risks of the odour control processes, which one might not be aware of. Moreover, some solutions to the problems were discussed.

The contaminants adsorbed in activated carbon can be released into the atmosphere, and spent activated carbon can become hazardous wastes. Moreover, during the regeneration of the spent activated carbon, greenhouse gas and water pollution can be produced. The chemicals used in the chemical scrubber can lead to climate change, freshwater eutrophication, photochemical oxidant formation, human toxicity, and ecotoxicity. The thermal treatment consumes a lot of energy, produces secondary emissions such as nitrous and sulphur oxides, and causes photochemical reaction. It also may produce undesired by-products, such as Cl_2 and DLCs. The main potential problems of biological treatment include the emissions of bioaerosol and the acidification and eutrophication of water. The high toxicity by-product formed during the UV photodegradation should be considered carefully. The formation of O_3 , NO_x , other VOCs, and aerosols during the non-thermal plasma should be paid attention to.

Process control and optimization are very important to solve these potential problems. Moreover, combinations of different technologies can be an attractive approach to solve the potential problems and improve the efficiency of odour control.

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