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N_2O emissions from full-scale nitrifying biofilters

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ABSTRACT

A full-scale nitrifying biofilter was continuously monitored during two measurement periods (September 2014; February 2015) during which both gaseous and liquid N_2O fluxes were monitored on-line. The results showed diurnal and seasonal variations of N_2O emissions. A statistical model was run to determine the main operational parameters governing N_2O emissions. Modification of the distribution between the gas phase and the liquid phase was observed related to the effects of temperature and aeration flow on the volumetric mass transfer coefficient (k_La). With similar nitrification performance values, the N_2O emission factor was twice as high during the winter campaign. The increase in N_2O emissions in winter was correlated to higher effluent nitrite concentrations and suspected increased biofilm thickness.

1. Introduction

Wastewater treatment using immersed biofilters combines both physical and biological processes. The benefits of this technology lie in its compactness, its small foot print, and intensiveness, i.e. its short residence time. This type of wastewater treatment reduces reactor size by 70% compared to conventional activated sludge systems (Rocher et al., 2012) and can be implemented both for carbon and nutrient removal. Biofilters are therefore an alternative to activated sludge tanks because they perfectly meet the effluent quality requirements required for large urbanized areas where building pressure makes land availability scarce (Tchobanoglous et al., 2003). Biofiltration is the most widely applied technology in the Paris area and the second-ranked technology in France after activated sludge (estimation based on analysis of nominal EP load of the French Ministry of Ecology database: http://assainissement.developpement-durable.gouv.fr/services.php).

Nitrous oxide (N_2O), which has a mean residence time of 120 years in the troposphere combined with a considerable ozone-destroying capacity in the stratosphere, has a global warming potential 265-fold greater than carbon dioxide on a 100-year horizon (IPCC, 2013; Ravishankara et al., 2009). Overall, N_2O contributes 6–8% of the anthropogenic green-house effect, despite its relatively low atmospheric concentration (322 ppbv) (IPCC, 2014; Montzka et al., 2011). In wastewater treatment plants (WWTPs) the release of N_2O from nitrification and denitrification processes has been clearly recognized (Kampschreur et al., 2009; Law et al., 2012b). Recent studies have suggested that ammonium oxidizing bacteria (AOBs) rather than heterotrophs could be the main source of N_2O production in WWTPs (Colliver and Stephenson, 2000; Guo, 2014; Wunderlin et al., 2012). Until now, research has focused on monitoring nitrous oxide emissions on activated sludge WWTPs and little is known about emissions from biofilm processes. However, recent modelling results have suggested that N_2O emissions from nitrifying biofilms could be significantly higher than those from suspended growth systems under similar conditions (Sabba et al., 2015). Since biofiltration technology is widely used, accurate quantification of its greenhouse gas emissions is required.

An overview of full-scale measurements indicates that the fraction of the inlet nitrogen load emitted as N_2O varies from 0 to 25% (Law et al., 2012a). Recent long-term monitoring studies reported average emission factors (EFs) of 0.12% (Rodriguez-Caballero et al., 2014) and 2.80% (Daelman et al., 2015) and shed light on the

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high temporal variability of the emissions. This discrepancy in the published data is probably due to differences in plant configurations and operational conditions. The methodology used for quantifying emissions has also been identified as a source of variability (Daelman et al., 2013).

During nitrification, two major biochemical pathways are involved in N_2O production by AOBs (Wunderlin et al., 2012). In the first pathway, N_2O is generated as a by-product of incomplete oxidation of hydroxylamine (NH_2OH) to NO_2, either through chemical decomposition of nitroxyl radical (NOH) or biological reduction of nitric oxide (NO). In the second pathway – so-called nitrifier denitrification – N_2O is generated upon the reduction of nitrites. A low dissolved oxygen concentration, nitrite accumulation and a sudden increase in nitrogen load have been identified as the main parameters associated with N_2O emission (Ahn et al., 2010; Foley et al., 2010; Kampschreur et al., 2009; Tallec et al., 2006a). A low dissolved concentration associated with insufficient air supply is likely to cause nitrite accumulation in the liquid phase and promote N_2O production through nitrifier denitrification (Colliver and Stephenson, 2000; Kampschreur et al., 2009; Tallec et al., 2006a). An increased nitrogen load is suspected of promoting a shift in metabolism from a low hydroxylamine oxidation activity towards the maximum activity inducing higher N_2O emissions (Chandran et al., 2011; Yu et al., 2010). In addition to environmental parameters, such as pH and temperature, transient shifts in operating conditions have been identified as promoting N_2O production (Chandran et al., 2011; Rodriguez-Caballero et al., 2014). The latter factor could be explained by either a disturbance of bacterial metabolism or the fact that such transitions will lead to a build-up of N_2O production pathway intermediates such as nitrite, hydroxylamine and ammonium.

The present study is the first monitoring campaign of N_2O emissions from a full-scale nitrifying biofiltration WWTP. Emissions were monitored during two periods in order to investigate seasonal variability. The main objectives were: (i) to provide the order of magnitude of N_2O EFs for nitrifying biofilters and (ii) to evaluate the temporal variability of the emissions in relation to process operating conditions. The ultimate goal is to provide the practitioners with validated data to better assess biofilters performances taking into account greenhouse gas emissions.

2. Material and methods

2.1. Description of the wastewater treatment plant

The measurements were taken at the Seine Aval WWTP, located in the Paris area in France. The plant was designed to receive a nominal flow of 1,700,000 m^3/d (around 20 m^3/s), i.e. about 80% of the Paris wastewater flow corresponding to about 5 million population equivalents. Wastewater is pre-treated (screening, grit removal) and passes through primary settling tanks. The water from the primary settlers enters a high loaded conventional activated sludge treatment (aerated biological reactors combined with secondary settling tanks) designed for organic carbon removal (Food to Microorganism ratio of 0.4–0.6 kgBOD/kgMLVSS/d). The outlet of the activated sludge system enters a ballasted flocculation unit to mainly remove suspended solids and phosphorus. Finally, the treated wastewater containing a low total suspended solid concentration and controlled phosphorus concentration is totally nitrified and denitrified through biofilters: 84 Biostyr® filters for nitrification followed by 18 Biostyr® and 12 Biofor® filters for post-denitrification, using methanol as an external carbon source.

Each nitrifying biofilter unit has a surface area of 173 m^2 and a total volume of 605 m^3. Microbial support material, composed of 4-mm-diameter polystyrene Biostyrene® spheres is distributed on the total biofilter surface area over a maximum height of 3.5 m. Nitrifying biofilters are continuously aerated through perforated tubes located under the support material. The aeration flow rate is automatically controlled on the 14-Biofilters-Pack outer ammonium concentration. The biofilter washing procedure is automatically triggered when material clogging reaches a given setpoint value. Washing consists of alternate injections of air and water in the filter bed, which removes the excess biomass. The washing procedure is carried out every 20–30 h and its duration is around 30 min.

2.2. N_2O sampling and monitoring

Two measurement campaigns were conducted on the same nitrifying biofilter unit to evaluate N_2O emissions and identify the key influential parameters. The first monitoring campaign – a summer campaign – was conducted in September 2014 and lasted 1 week. The second one – a winter campaign – was conducted in late January, early February 2015 and lasted 2 weeks. During both campaigns, liquid and gaseous emissions were monitored continuously.

Dedicated onsite experiments were also conducted during the winter campaign to further investigate the impact of the aeration pattern on gaseous emissions (during day 9 from 10 a.m. to 6 p.m.). To this specific aim, four aeration flow rates were successively imposed and maintained for at least 2 h. The air flow rate was applied to fit a fixed air load (60, 80, 100 and 120 Nm^3/kgN) which was calculated according to inlet NH_4 concentration and liquid flow at the beginning of each test.

After the summer campaign, a maintenance operation allowed the measurement of the support material height. A value of 2.95 m was measured, which is below the design value of 3.5 m. This variation is due to typical material loss during the filtration operation. After the campaign, the support material was refilled and its height was estimated to be 3.35 m during the winter campaign considering the 3-month loss of material.

The measurement and sampling points to calculate the gaseous and dissolved N_2O emissions are presented in Fig. 1.

The dissolved N_2O concentration was measured on-line through N_2O microsensors (Unisense A/S, N_2O-R, Denmark). A daily two-point calibration was systematically performed to ensure measurement error below 10%. Fresh distilled water was used for the zero point calibration, whereas a fresh N_2O concentration solution was prepared for the second point calibration. This solution was obtained bubbling a 500-ppmv standard gas in fresh distilled water. The resulting dissolved N_2O concentration was calculated according to Henry's law constant and temperature.

Gaseous samples were collected using a wooden floating hood, with a surface area of 1.6 m^2, and located in the middle of the reactor surface. This location was chosen because measurement of gas flow rate at different locations of the studied biofilter (performed prior to the first monitoring campaign, data not shown) indicated that in that position the local gas flow rate was close to the insufflated air flow rate. The gas flow coming out from the hood was measured using a precision flowmeter (System instrumentation, DMT586b, France) with a measurement range of 2.5–150 Nm^3/h. Surface gas flow measured during campaigns ranged from 7 to 30 $Nm^3/h/m^2$. A portion of the off-gas was directed to an infra-red analyzer (AP2E, ProCeas, France) to measure its N_2O concentration on-line. The analyzer has a measurement range of 0–55 ppmv and a detection limit of 50 ppbv. Calibration was performed once a week using N_2O gas standards. Since N_2O concentration in the exhausted gas was higher than the analyzer's upper measurement limit, dilution of the gas – with pure N_2 gas – was performed using two precision flow controllers