Measuring nitrous oxide emissions from biological wastewater treatment, art or science?

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Introduction

In light of sustainability in the water sector and the high global warming potential of nitrous oxide (N_2O), an intermediate in biological nitrogen removal, its emission from wastewater treatment plants (WWTPs) has been increasingly studied in recent years. Field measurements of N_2O emissions from WWTPs have subsequently been carried out for a number of different campaigns across many different parts of the world, employing several different methods, seemingly giving the measurements an artistic quality. The majority of these campaigns are actually research related, and their objectives vary from understanding potential emissions under different WWTP conditions (Daelman et al., 2013; Foley et al., 2010), to mechanistic modelling of the N_2O production and emissions from full-scale WWTPs (Ahn et. al., 2010; Guo et al., 2013). The latter objective, which is one of the main objectives of IWA Task Group on GHG modelling (Task Group GHG), requires a minimum level of full-scale data, and several datasets for gaining consensus on N_2O model validity. This becomes difficult when there are gaps in the data, as well as inconsistencies with how the data is collected and expressed. Although a comprehensive Water Environment Research Foundation (WERF) N_2O field measurement protocol (Chandran, 2009) exists, and is designed to meet the objectives of mechanistic modelling, it does not provide guidance as to potential alternative methods for meeting the same or similar objectives, where flexibility in the protocol can be afforded. Therefore, the aim of the Task Group GHG and this paper is to identify the areas in which additional guidance can be provided for researchers and practitioners interested in measuring WWTP N_2O emission for: a) obtaining a fairly accurate estimate of the magnitude of emissions for a given WWTP, and for a given set of conditions (operation, load, temperature); and b) calibrating and validating N_2O production models to understand potential pathways and be able to accurately describe observed production and emissions. The final guide should also help provide a minimum level of consistency in field data collection, such that the overall performance of different N_2O models can be fairly assessed when subjecting them to different field datasets. Finally a discussion on the way that N_2O emission data should be provided to ease the comparison between different studies is presented.

Methods

To identify the areas where additional guidance can be provided to complement what is already covered in the WERF Protocol, the N_2O field measurements guide development first consisted of reviewing both the WERF Protocol in detail and literature. Then, discussions within the IWA Task Group GHG network were initiated between members experienced in several N_2O measurement campaigns to identify, from experience, where allowances could be made to the WERF Protocol, and how emission factors could be expressed more consistently. An experimental campaign was also carried out by some of the Task Group members at a full-scale WWTP in the Netherlands (Aarle-Rixtel WWTP, Waterboard Aa en Maas). Gas hood types and hood placement within biological reactors were explored to measure off-gas N_2O emissions, and understand potential differences that need to be taken into account in hood selection and placement. Two hood types, a large hood and a small hood, were placed side by side to measure online N_2O gas concentrations with an off-gas N_2O analyzer (Emerson Rosemount X-STREAM Enhanced General Purpose Process Gas Analyzer), with automated valves for performing online measurements from multiple locations. The large hood was built by the BIOMATH Department at Ghent University for measuring oxygen transfer (ASCE, 1996; Rosso et al., 2005). The small hood is the US EPA surface emission isolation flux chamber (SEIFC) type. To examine potential N_2O dynamics across the biological treatment train, two of the SEIFC flux chambers were placed in two adjacent aeration zones, with no anoxic zones in between, to measure online N_2O gas concentrations from the two locations (approximately 20 meters apart) simultaneously.

Results and Discussion

Reviewing both the WERF Protocol and literature resulted in identifying the following areas where flexibility in the WERF Protocol could be afforded and guidance provided for alternative methods: 1) Process data collection and review prior to measurements; 2) Minimum liquid and process data requirements during measurements; 3) Sampling hood / flux chamber types; 4) Flux determination; and 5) Gas and liquid N_2O analysis techniques and equipment. Therefore, the proposed N_2O modelling field measurements guide will address these areas and provide specific guidance on potential alternatives to the protocol. For example, the WERF protocol describes a helium tracer method for determining N_2O flux, however, there have been other successful methods, which may offer a solution in cases where the helium tracer method may not be feasible.

Regarding experimental results investigating hood types, Figure 1A shows two different hood types placed side by side to examine hood design impacts on measurements, while Figure 1B shows the concentrations measured by each. Both hood types exhibited very similar concentrations and dynamics; however, the large hood at times measured slightly higher concentrations, by approximately 3 ppmv. This could be due to pressure build up in the large hood, causing an accumulation of gas, as it did not have a vent port. The smaller SEIFC hood is designed with a small venting port to prevent pressure build up. Given the larger surface area of the large hood, this pressure build up may not be as pronounced as in a smaller hood not having the vent. This highlights the benefit of having a vent similar to the SEIFC flux chamber regardless of hood size. These results also indicate that hood size should not significantly impact measurements assuming they are properly vented. Of course, the larger surface area will provide better averaging of emissions in a given zone if there is significant spatial variability in a given zone since it provides greater surface area to capture emissions. This will be discussed in detail in the proposed guidance.

Figure 2 illustrates the potential variability in emissions across the biological treatment train for a given condition as the two SEIFC hoods were used to measure emissions from the two locations (end of Zone 1 and beginning of Zone 2) at the same time. Both locations had the same diffuser density and received air from the same air header without throttling between the header and diffusers. Both hoods exhibited very similar concentrations and dynamics; however, the first hood, at the end of Zone 1, exhibited slightly higher emissions concentrations, by approximately 4 ppmv in some cases. The upstream hood may have had higher concentrations due to likely higher ammonia concentrations, different dissolved oxygen (DO) concentrations; hence different ammonia oxidation rates and N_2O production (Law et al., 2012). Another possibility could be due to different nitrite (NO_2^-) concentrations from different DO concentrations (Hanaki et al.,1990; Mota et al., 2005); hence, different N_2O production from ammonia oxidizing bacteria (AOB) denitrification (Kampschreur et al., 2009). From this experience we have learned that monitoring these additional parameters (NO_2^-, DO, and NH_4^+) at each location at the same time, along with liquid grab samples of dissolved N_2O, would help confirm which of these possibilities / mechanisms are more likely when comparing simultaneous measurements from different locations. Guidance on this will be provided in the proposed measurements guide.

Figure 2 also indicates that there is significant N_2O production within the adjacent aerobic zones, as confirmed by similar concentrations and peaks measured in both upstream and downstream aerobic locations. Since there was not a large difference in concentration between the two locations, the results indicate that one may opt to only measure in one location within adjacent aerobic zones of a similar WWTP configuration and air piping, if only trying to gain a reasonable estimate of the emissions. However, if trying to understand and describe mechanistic behavior through modelling, then measuring N_2O in multiple locations simultaneously, along with the other parameters suggested above, would help in gaining a mechanistic understanding of the emissions, and confirm whether or not there are significant differences in concentrations/emissions across the treatment train. Clarification on what approaches are appropriate for different objectives, such as the above, will be included in the proposed guidance.

Regarding emission factors, it is recommended to express N_2O emitted per nitrogen converted rather than N_2O emitted per nitrogen load, because N_2O production will only come from the transformation of the influent nitrogen, and not from the nitrogen load. If data is expressed as N_2O emitted per nitrogen load, then the percent of nitrogen removed or converted in the system should also be expressed. However, the type of system should also be considered. For example, expressing N_2O emission by removed load may make less sense for a one-stage partial nitritation-anammox system, where you may not know exactly how much ammonium was converted by ammonia oxidizing bacteria and how much by anammox (the latter not producing N_2O). In such case, it may be better to express the N_2O emission per ammonium influent load. The discussion on emission factors will be detailed in the proposed guidance.

Conclusions

The new guidance on measuring N_2O emissions from full-scale WWTPs, proposed by the IWA Task Group GHG, will provide greater flexibility for monitoring N_2O, while ensuring a rigorous assessment of those emissions. In this paper, various aspects of measuring N_2O emissions from full-scale WWTPS have been identified as needing guidance on alternatives. For example, hood types were examined to see the effect that hood types or locations can have in the data collected. The results showed that, depending upon objectives, there are different considerations that should be taken into account by operators and/or scientist when

developing measurement plans. The IWA Task Group GHG has also established recommendations on the way that N_2O emission factors are being presented to facilitate comparisons between different studies. The full guide on N_2O Field Measurements for WWTPs will provide a flexible consensus-based approach for measuring sustainable biological nitrogen removal in WWTPs.

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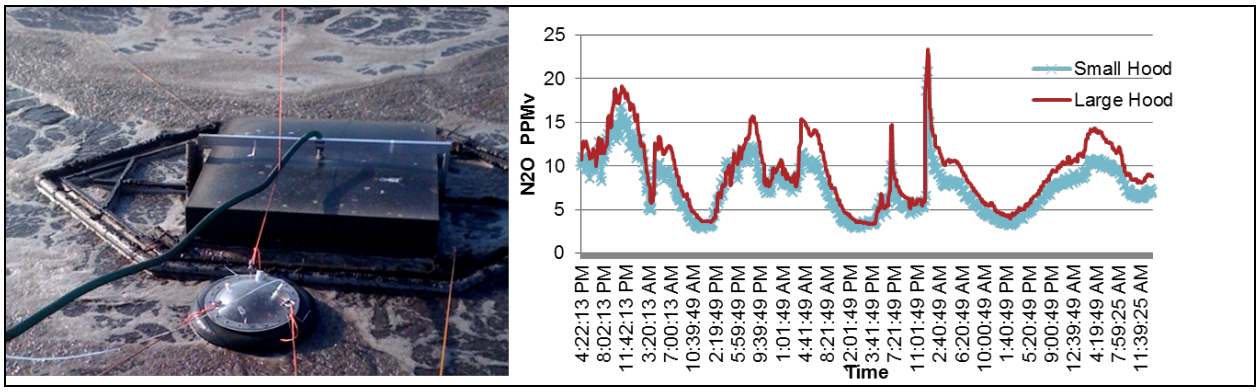

A **B**

Figure 1 – Comparison of N_2O concentrations measured in different hood types

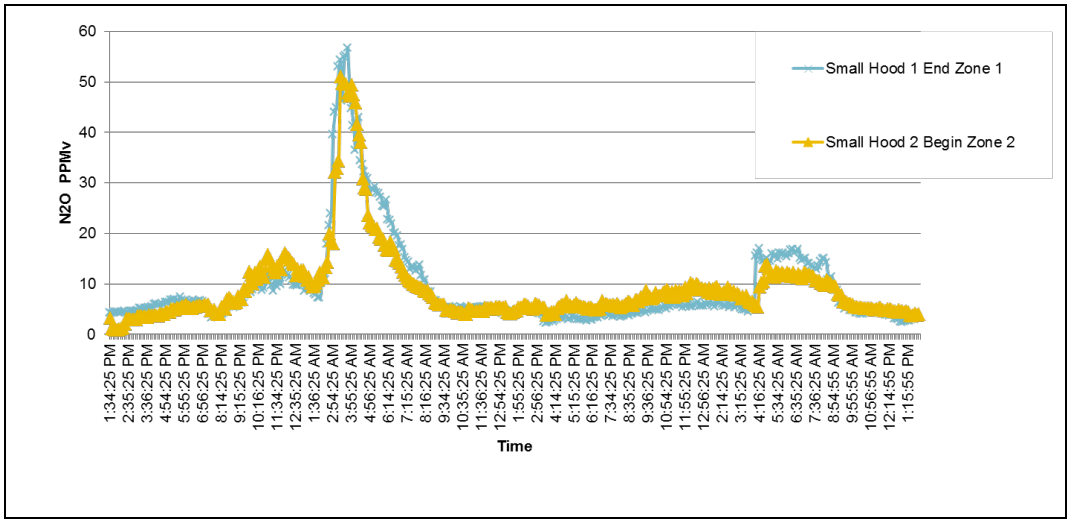

Figure 2 – Comparison of simultaneous measurements with same hood type in different locations