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APPROACH FOR A NOVEL CONTROL STRATEGY FOR SIMULTANEOUS NITRIFICATION/DENITRIFICATION IN ACTIVATED SLUDGE REACTORS

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Abstract—Biological nitrogen removal in activated sludge processes is conventionally obtained by a sequence of aerobic and anoxic processes. Kinetic mechanisms affecting the oxygen balance could trigger the production of total volatile organic carbon (TVOC) and nitric oxides (NO) under anoxic and anaerobic conditions. Measurements at a wastewater treatment pilot plant of capacity 1.6 m^3 wastewater show that the amount of TVOC and NO produced during the treatment process depends on carbon loading (low feed, balanced and overloaded) and aeration conditions. To confirm the results ORP, pH and dissolved oxygen (DO) are measured on-line and chemical parameters such as nitrate (NO_3-N), ammonium (NH_4-N) and TOC are measured in the wastewater.

The ORP observed is in the range of −60 and +198 mV includes optimal setpoints for simultaneous nitrification and denitrification. From the pattern of NO emission plot, it can be assumed that a part of the produced NO_3-N is denitrified during the aeration period. NO emissions are especially high during denitrification conditions at low oxygen rates. The results suggest that both NO and TVOC emission concentrations in combination with ORP can be valuable parameters to control operation of a wastewater treatment plant. Continuous measurements of ORP and NO concentrations for estimation of NO emissions gauging the extent of nitrification or denitrification in the plant becomes possible.

Key words—denitrification control, nitric oxides, total volatile organic carbon, oxidation-reduction potential

INTRODUCTION

Biological nitrogen removal in activated sludge processes is conventionally obtained by a sequence of aerobic and anoxic processes. Normally, for simultaneous denitrification, carbon is finally degraded to carbon dioxide, CO_2, and the remaining available dissolved oxygen, DO, can provide nitrifying conditions. NO_3-N containing effluent can be either recirculated and mixed with the raw sewage, or denitrified by adding external carbon sources like methanol, ethanol or acetate (Münch *et al.*, 1996; Isaacs *et al.*, 1994; Isaacs and Henze, 1995).

Removal of nitrogen from wastewater is an essential regulation in several countries of the European Union. In Austria, nitrogen removal rate of 60–70% (BGBl 210/1996) and a decrease of NH_4-N to 10 mg l^{-1} and NO_3-N in wastewater is necessary.

The monitoring of nitrogen removal can be performed by photometric or by electrode-measurements of NO_3-N and NH_4-N. For the control of nitrification/denitrification these parameters are less useful because real-time control is difficult to achieve.

According to Lie and Welander (1994) complete denitrification is obtained in the absence of DO. DO was found to have a negative effect on denitrification even at lower concentrations that can be measured with conventional DO probes (i.e. <0.1 mg l^{-1}) (Lie and Welander, 1994). The oxidation-reduction potential, ORP, was found to be a useful indicator of the DO concentration at this low level and the denitrification rate was found to decrease linearly with increasing ORP. However, there are also limitations in the use of ORP for the control of denitrification (Csikor *et al.*, 1996). Al-Ghusain *et al.* (1994) express scepticism of ORP measurements and state the need for better parameters such as pH. There remains a need for control of parameters for optimised DO supply for

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nitrification/denitrification conditions. Kinetic mechanisms affecting the oxygen balance have been shown to trigger the production of nitric oxides, NO, especially under anoxic and anaerobic conditions (Schulthess *et al.*, 1994). According to Schulthess *et al.* (1994) nitrous gases are produced at DO concentrations of more than 0.5 mg l^{-1} O_2. Under anoxic conditions, when the oxygen supply drops to zero—especially during the denitrification process—the concentration of nitric oxides increases.

OBJECTIVES

The objective of this work is to characterise the emission rates of nitric oxide (NO), total volatile organic carbon (TVOC), and CO_2 in the off-gas emissions of wastewater treatment plants under nitrifying and denitrifying conditions in order to assess their usability as control parameters for optimised oxygen supply. The aeration rate is kept at different DO levels while monitoring NO in the gas phase as NO is produced under low oxygen/denitrifying conditions. The approach is to show the impact of different aeration times and oxygenation rates in the wastewater on NO production. This study is intended to improve the understanding of the NO production as well as TVOC forming effects under different ORP, DO and carbon loading conditions (low-feed, balanced and overloaded conditions).

MATERIALS AND METHODS

The investigations are carried out in an activated sludge unit of a wastewater treatment pilot plant. Figure 1 shows a schematic of the reactor used to perform the experiments. The polypropylene activated sludge plant with a water capacity of 1.6 m^3 is fed with fresh Viennese wastewater. The time controlled feed is delivered during aeration and non-aeration periods at a rate of 4.4 l min^{-1} and the feed periods are chosen to be 1.5 min on and 4 min off, and 3 min on and 4 min off, producing influent rates of 1.7 m^3 day^{-1} and 2.7 m^3 day^{-1}, respectively. These flows give hydraulic retention times (HRT) of 25 h and 14 h, respectively. Aerobic treatment, nitrification and denitrification are carried out in the same reactor.

The mixed liquor suspended solids concentration in the aeration tank is in the range of 4 g l^{-1} in Stage I, 5 g l^{-1} in Stage II and 7 g l^{-1} in Stage III. The sludge recycling rate is constant and about 1.7 m^3 day^{-1}. Wastewater temperatures during investigation periods ranged from 18 to 19°C. Time-controlled aerators aerate the wastewater at a rate of 16 Nm^3 h^{-1} (Nm^3 are based on 0°C and 1013 mbar). Additional air (5.5 Nm^3 h^{-1}) is pumped continuously into the headspace of the airtight chamber to maintain the air flow during non-aeration times and to avoid the collapse of the cover.

Off-gas samples are collected from the outlet of the airtight chamber through a glass manifold which is kept at 25°C. In the off-gas NO/NO_x, CO_2 and TVOC are measured. Volatile compounds such as NO/NO_x and TVOC and CO_2 are examined with continuous automatic facilities (Nitrogen Oxides Analyzer Model 8840, Monitor Labs, a TOC-Analyser Model VE with FID, Messer Griesheim and UNOR 6N NDIR, Maihak). Comparing NO and NO_x measurements, results for NO_x are in the same range as NO. However, due to quenching effects the NO_x concentration is about 10% smaller than that of NO. The TVOC is measured as total combustible organic vapour content as propane equivalent, and the concentrations and emission rates are calculated in mg Nm^{-3} carbon or mg min^{-1} carbon. Influent samples are taken after the storage tank and the effluent samples are taken at the outlet of the aeration chamber prior to the settling tank. The excess sludge of the treatment plant is removed with the effluent. To obtain a rough idea of the performance of the WTP, samples are investigated after sludge removal. Samples are analysed for total organic carbon (TOC), dissolved or-

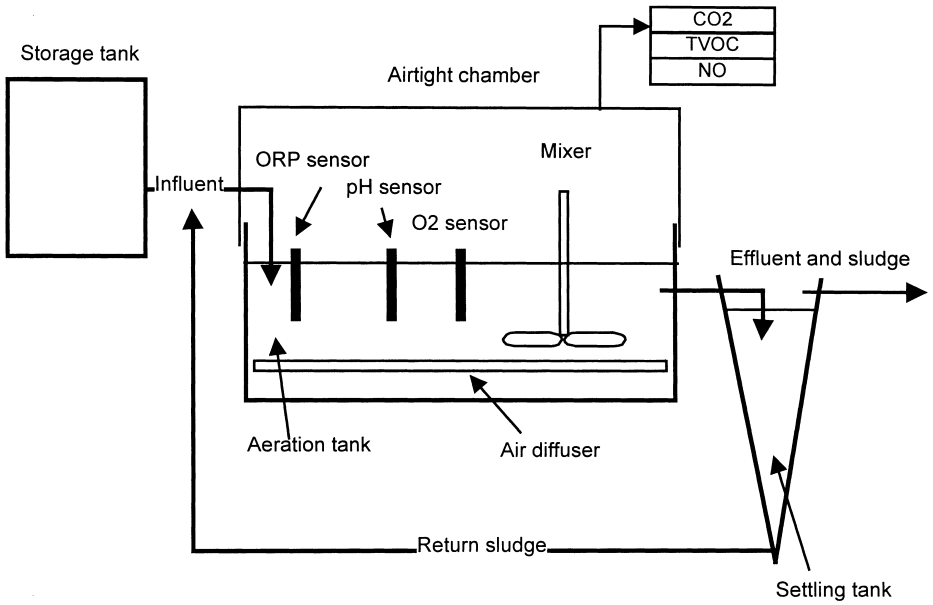

Fig. 1. Schematic diagram of the treatment plant.

Table 1. Average concentrations of the influent during Stage I to III

TOC (mg l^{-1})	NH_4-N (mg l^{-1})	N-org. (mg l^{-1})	PO_4-P (mg l^{-1})	P-total (mg l^{-1})	SS (mg l^{-1})
159 ± 29	48 ± 5	24 ± 6	3.0 ± 1.2	5.3 ± 2.4	150 ± 29

ganic carbon (DOC), total Kjeldahl Nitrogen (TKN), phosphate (PO_4-P), total phosphorus (P_{total}), suspended solids (SS), NH_4-N and NO_3-N. The analytical procedures follow the DIN (German Industrial Standards) analytical methods (DEV, 1998).

RESULTS

Three different stages are considered to show the response of the activated sludge system in terms of NO, TVOC and CO_2-production. System circumstances include Stage I with low load and high aeration, Stage II with balanced feed and aeration and Stage III under overloaded conditions. Table 1 shows the average concentration of wastewater quality in the influent during experimental periods.

The operating conditions and the concentrations of TOC, NH_4-N and NO_3-N in the influent and effluent, and the minimum and maximum DO and ORP in the liquid phase of the aeration tank at different stages are presented in Table 2.

Minimum and maximum concentrations of TVOC, CO_2 and NO in the off-gas of the aeration tank at different stages are given in Table 3. Figures 2–7 include the emission rates of CO_2 in mg min^{-1} C, NO in mg min^{-1} and TVOC in g min^{-1} C, as well as the changes in DO concentration as O_2, pH and ORP during Stages I–III. All recorded periods start with the aeration cycle. As the flowrates of air changed with aeration and non-aeration time, TVOC, CO_2 and NO in the off-gas is calculated in mg min^{-1}.

Stage I

The influent concentration of TOC was considerably low when Stage I was conducted (Table 2). During low feed periods with high aeration times, high TOC removal from wastewater is expected. The CO_2 emission rate rises sharply with the beginning of aeration (Fig. 2). After reaching the peak stage of CO_2 production, the concentration is diluted by the additional air in the airtight chamber. Short breakpoints can be obtained because degradable substances are added with the raw sewage during aeration and non-aeration periods. The removal of wastewater TOC is about 170% of the total influent load of 218 g day^{-1} TOC based on CO_2 production; with a residual of 7.6 mg l^{-1} dissolved organic carbon (DOC) in the liquid phase. This means that sludge has been degraded under this condition; the calculated loss is about 2.3% of the sludge in the aeration tank per day. The ORP in Stage I remains at quite high mV-ranges between 115 and 290 mV, and there is no major breakpoint of denitrification visible. The corresponding DO levels are quite high and reach high levels of 5 mg l^{-1} O_2 (Fig. 3).

As raw sewage is added continuously during various intervals, denitrification takes place simultaneously along with nitrification during aeration. Nearly whole of NH_4-N is metabolised and well nitrified in Stage I. The remaining NH_4-N concentration in the effluent is due to the addition of fresh wastewater during non-aeration. A major portion of NO resulting from previous denitrification under anoxic conditions is stripped at the beginning of the next aeration cycle. In Fig. 2, the NO-emission rate rises sharply due to the stripping effect but decreases during the first part of the aeration and the stripping is quickly over as seen in the sharp decrease of the NO emission rate (see Fig. 2). The initial increase in NO emissions shows the NO production during the non-aeration phase, though the nitrate-breakpoint in the ORP-graph is not visible. The average NO_3-N concentration of the effluent in Stage I is still 2.5 mg l^{-1} NO_3-N due to high and long aeration periods.

Table 2. Operating conditions, concentrations of TOC, NH_4-N and NO_3-N in the wastewater influent and effluent, and minimum and maximum values of ORP and DO in the wastewater

	Stage I	Stage II	Stage III
Aeration time (h)	2	0.5	0.5
Non aeration time (h)	1	1	1
Inflow (m^3 day^{-1})	1.7	1.7	2.7
TOC influent (mg l^{-1})	126	170	180
NH_4-N influent (mg l^{-1})	51.5	50	41.8
DOC effluent (mg l^{-1})	7.6	10	13
NH_4-N effluent (mg l^{-1})	0.8	1	25.3
NO_3-N effluent (mg l^{-1})	2.5	< 0.5	0.8
N-removal rate (%)	96	98	62
O_2 max (mg l^{-1})	5.4	1.3	0.2
O_2 min (mg l^{-1})	0	0	0
ORP max (mV)	290	198	100
ORP min (mV)	115	−33	−71
pH max	7.3	7.1	7.1
pH min	7.0	7.0	6.8

Stage II

For Stage II the changes in TVOC, CO_2 and NO emission rates are shown in Fig. 4. The CO_2 con-

Table 3. TVOC, CO_2 and NO in the off-gas of the aeration tank

	Stage I	Stage II	Stage III
TVOC max (mg m^{-3} C)	38.4	84.0	127
TVOC min (mg m^{-3} C)	11.7	24.4	35.2
CO_2 max (vol%)	0.27	0.32	0.39
CO_2 min (vol%)	0.07	0.10	0.11
NO max (ppbv)	91.8	91.3	109
NO min (ppbv)	34.7	52.3	57.6

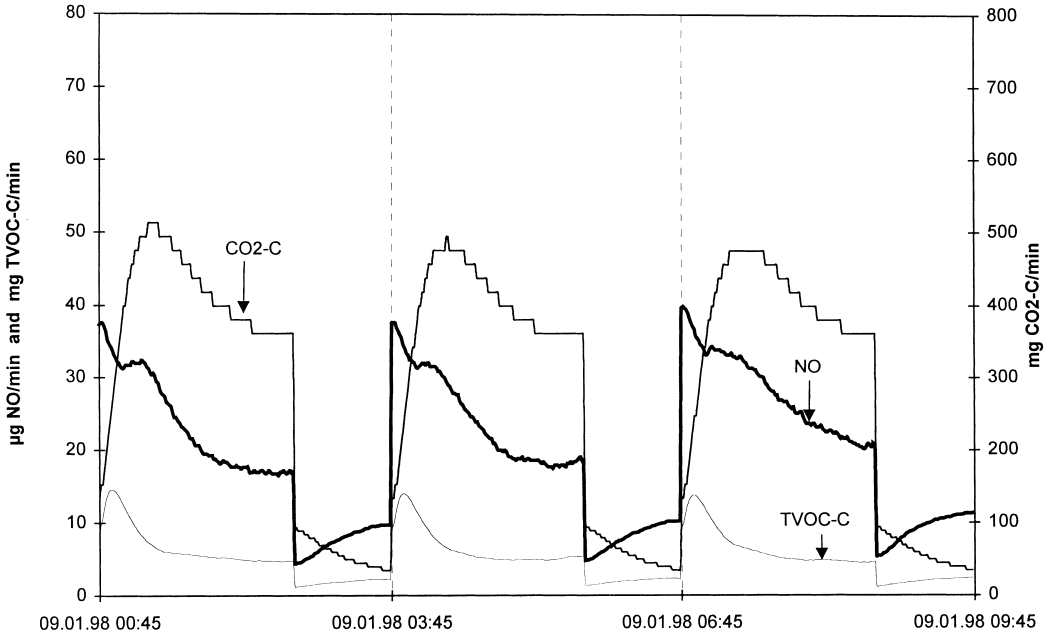

Fig. 2. Actual emissions of CO_2 in mg min^{-1} carbon, NO in μg min^{-1} and TVOC in mg min^{-1} carbon during Stage I (the interrupted lines indicate the start of the aeration periods).

centration increases during aeration and drops when the air supply is turned off. The final oxidation of TOC, based on CO_2 measurements, is 90% of the carbon inflow (294 g day^{-1} TOC). DOC measures 10 mg l^{-1} in the effluent, equalling 6% of the C in the influent. A rate smaller than 100% means that sludge is accumulated in the system. Stage II shows ORP-ranges between -33 and 198 mV and DO levels between zero and 1.3 mg l^{-1} O_2. Denitrification breakpoints can be seen quite clearly (Fig. 5).

In Stage II the removal of NH_4-N is similar to

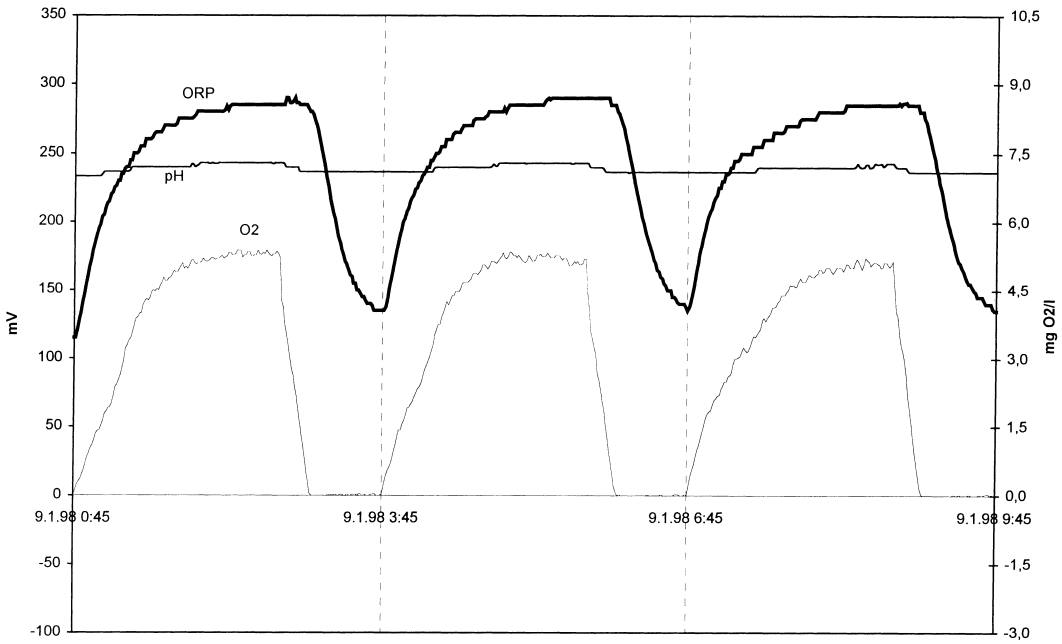

Fig. 3. Changes in DO concentration, pH and ORP during Stage I (the interrupted lines indicate the start of the aeration periods).

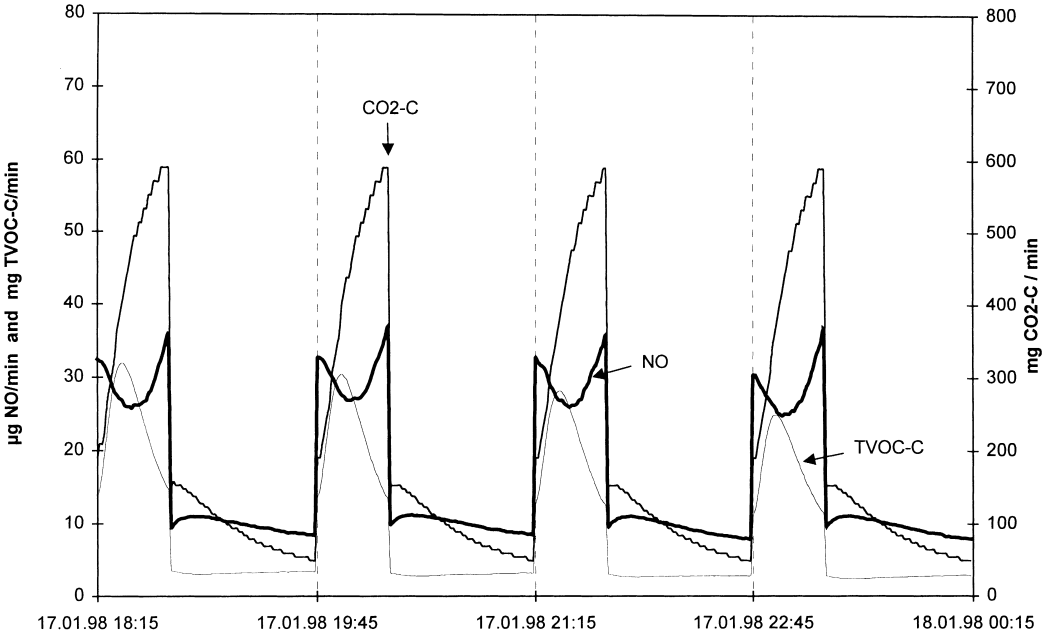

Fig. 4. Actual emissions of CO_2 in mg min^{-1} carbon, NO in μg min^{-1} and TVOC in mg min^{-1} carbon during Stage II (the interrupted lines indicate the start of the aeration periods).

that of Stage I. In this case the raw sewage also contributes to the NH_4-N concentration in the effluent. The plant is in a balanced stage according to TOC concentrations, and NO_3-N is removed to concentrations below detection limits. Under aerobic conditions the heterotrophic bacteria produce TVOC as an intermediate product and CO_2 as the final product. During the ascending stage of the TVOC graph there is not enough oxygen for the complete oxidation of the organic carbon and no oxygen left for nitrification. It is assumed that nitrification starts after the TVOC emissions have

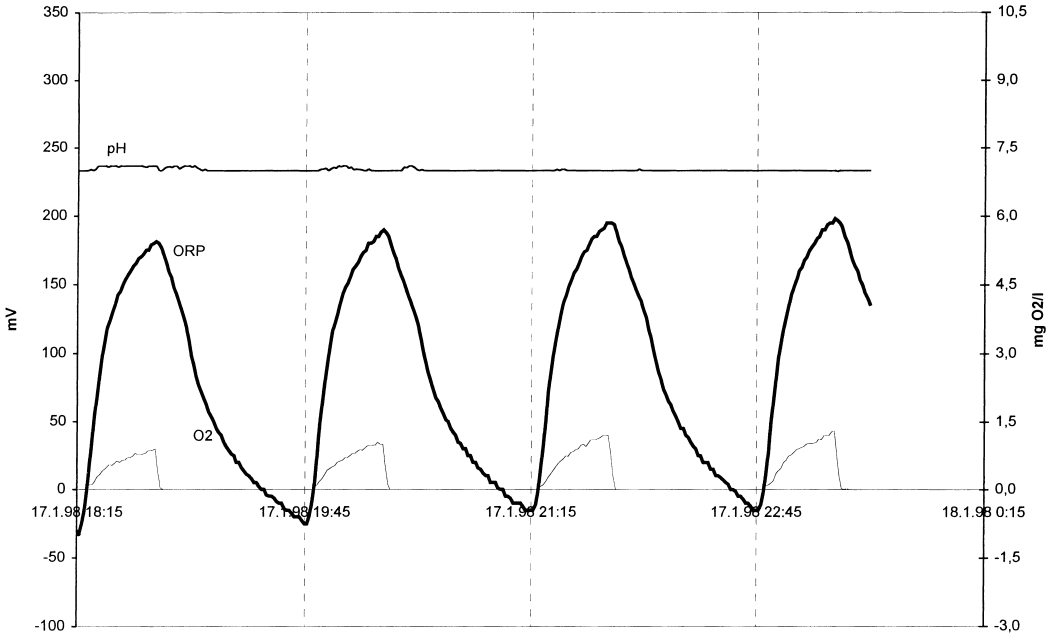

Fig. 5. Changes in DO concentration, pH and ORP during Stage II (the interrupted lines indicate the start of the aeration periods).

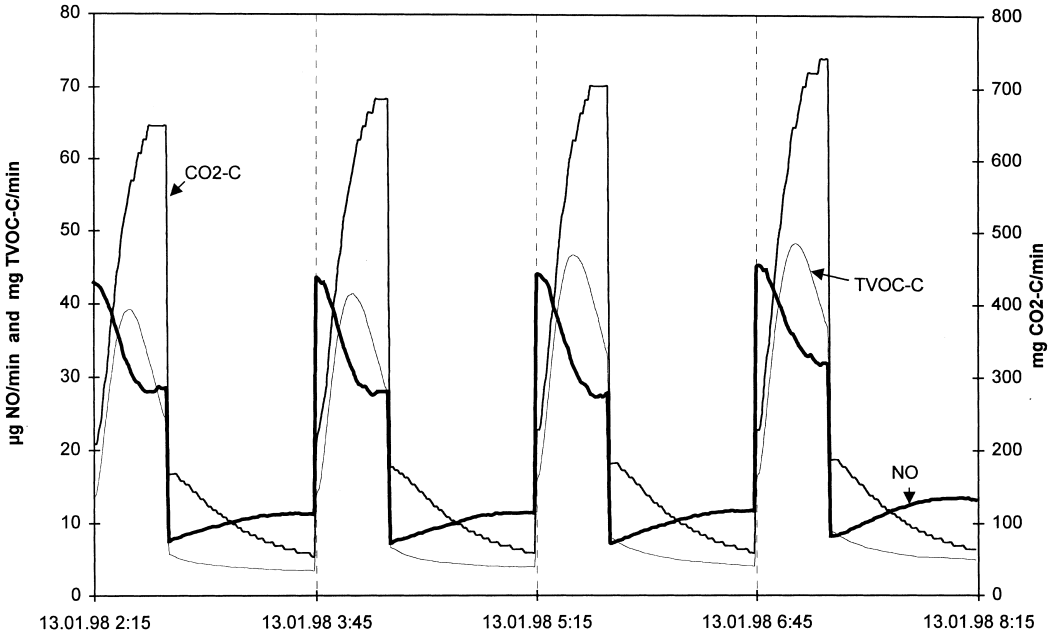

Fig. 6. Actual emissions of CO_2 in mg min^{-1} carbon, NO in μg min^{-1} and TVOC in mg min^{-1} carbon during Stage III (the interrupted lines indicate the start of the aeration periods).

passed the maximum and the CO_2 graph flattens. Here the NO emissions reach a minimum indicating that the NO emissions produced during the non aerated period have been stripped. The ascending graph indicates simultaneous denitrification since the NO is a by-product only of denitrification. Denitrification is also shown by the ORP graph.

Stage III

At the 2.7 m^3 day^{-1} influent rate in combination with short aeration periods the pilot plant is already overloaded. Thus carbon removal and nitrification are not complete. The changes of TVOC, CO_2 and NO loads are shown in Fig. 6. The increased TOC

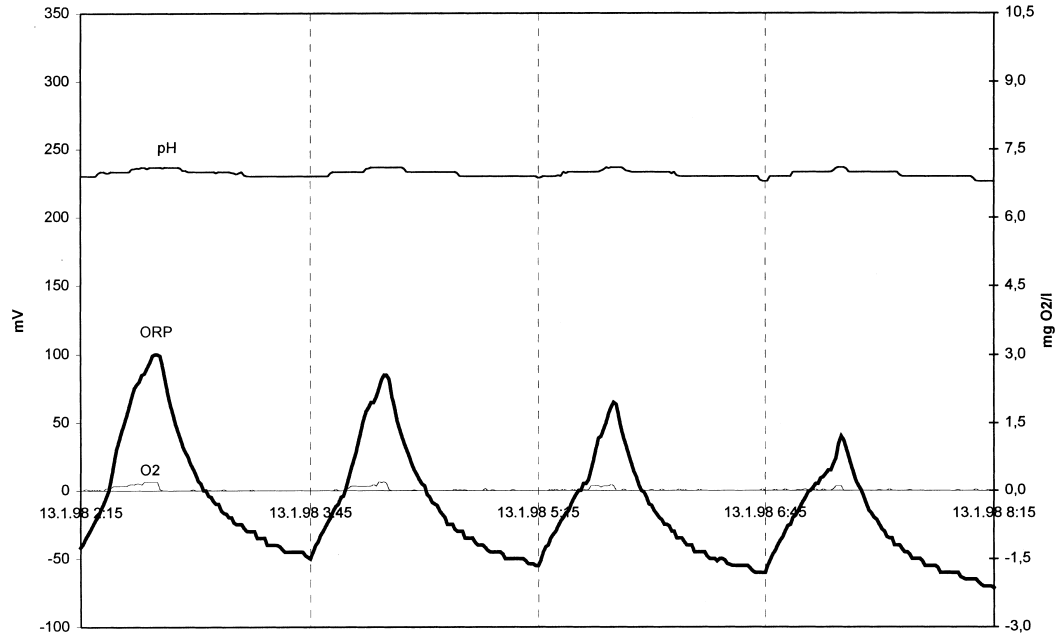

Fig. 7. Changes in DO concentration, pH and ORP during Stage III (the interrupted lines indicate the start of the aeration periods).

loads in the influent also lead to higher CO_2, as well as TVOC emission rates (see Fig. 6) since the oxidation cannot carry on with the carbon degradation. The CO_2 emission rate reaches its peak slowly and drops sharply after aeration stops. The elimination of TOC, based on the CO_2 investigation, is only 48% of the carbon in the influent (489 g day^{-1} C). Carbon in the effluent is 13 mg/l DOC (35 g day^{-1} C). Overloaded conditions produce additional TVOC during aeration (Fig. 6). Under these conditions the ORP-graph drops to −71 and 100 mV and keeps decreasing as the TOC loads rise. The O_2 concentration hardly reaches measurable values and behaves similarly to the ORP (Fig. 7). No denitrification breakpoints can be monitored. The variation in pH is as low as 0.2 and is quite neutral in all three stages.

Under the overload conditions of Stage III insufficient oxygen is available for full nitrification, although the performance of the plant is good for removal of dissolved carbon. The NO emission plot decreases less sharply and the TVOC emission rates are considerably higher than under low carbon load conditions. The TVOC and CO_2 plots (see Fig. 6) indicate that the air consumption is just enough to brake down organic carbon. Little O_2 is left for NO_3-N production at the end of the aeration period. In Stage III denitrification generates NO mainly during the non-aeration period and during the aeration period to a lesser extent than in Stage II. This is demonstrated by an increase in NO emission rates during non-aeration and by the stripped peak at the beginning of aeration.

DISCUSSION

Schulthess *et al.* (1994) investigated the net-production of the denitrification intermediates NO and nitrous oxide (N_2O) at various concentrations of DO in reactors for ranges from 0 to 4 g O_2 m^{-3}. They found that NO is produced by denitrifiers and that high NO_2-N in the wastewater as well as aerobic conditions favour the production of N_2O but not NO. Furthermore, NO is accumulated at very low concentrations and the highest emission rates occur under completely anoxic conditions.

The experiment reported here was carried out during three months and to demonstrate characteristic conditions the three extreme stages outlined were selected. To figure out changes in the off-air development unbalanced conditions including sludge accumulation and depletion in the system needed to be applied. Reproducible characteristic graphs of NO, TVOC and CO_2 emissions and of ORP and DO concentration were produced illustrating the changes in TVOC and NO concentrations during variations of oxidation time, feed rate and the influence on the ORP. The ORP variations rely mainly on the feeding rate and the concentration of the return sludge in the settling tank and also depend directly on the concentration of biodegradable carbon. High TVOC concentrations show that the carbon breakdown is not yet completed and that more oxygen is needed for carbon removal. NO production indicates the start of the nitrification and denitrification process.

ORP is a very complex parameter and depends on both the wastewater quality and the performance of the sludge. Consequently not only the height of the ORP is commonly used for aeration control, but also the breakpoints. Since the ORP has a broader range than DO it is better suited for process control for denitrification than DO. Even in anoxic conditions changes in ORP can be monitored but these points are not always detectable, even in nitrifying and denitrifying plants. The optimal nitrification and denitrification could be measured at ORP values between −33 and +290 mV in Stage I and Stage II. The optimal ORP setpoints for denitrification control are reported in a wide range and depend on the wastewater used. Lo *et al.* (1994) found setpoints for ORP between 40 and 180 mV with an optimum at 100 mV, whereas de-la-Menardiere *et al.* (1991) suggest 200 and 400 mV and Ukita *et al.* (1990) suggested 250 and 350 mV for carbon and nitrogen removal as lower and upper limits, respectively. The optimal setpoint for simultaneous nitrification-denitrification is given by Okamoto *et al.* (1990) at 125 mV. The observed ranges meet most of the given optimal ORP-ranges.

In contradiction to Lie and Welander (1994) who found that a major problem for denitrification is the inhibitory effect when traces of oxygen are present even below the detection limit of commonly used O_2-electrodes (detection limit 0.1 mg l^{-1}), this study demonstrated that fairly anoxic conditions are fit to oxidise and reduce nitrogen simultaneously. Neither short-term experimental anoxic conditions nor low ORP inhibited the activity of the nitrifying–denitrifying capacity. The denitrification effect is indicated by the NO production and monitored by analysing NO_3-N and NH_4-N.

The effect of the ORP on denitrification differs from plant to plant (Lie and Welander, 1994) and the control of denitrification by monitoring of the breakpoint is difficult. Therefore a good possibility for the process control is to measure the NO production in the off-gas of the aeration tank of wastewater treatment plants. These data, plus time plots of ORP, DO-concentration, CO_2 and chemical analyses demonstrate that oxygen is being actively consumed by both carbon and nitrogen oxidisers and that denitrification is detectable by real time measurements of NO emissions. With the monitored parameters (CO_2, DO, NO, TVOC and ORP) the oxygen supply of treatment plants could be controlled and optimised.

CONCLUSIONS AND FUTURE ASPECTS

The process of biological nitrogen removal in one reactor with nitrification and denitrification taking place in series in the same tank, was monitored by measuring the parameters ORP, pH and DO in the wastewater in combination with TVOC, CO_2 and NO in the off-gas. This provided the possibility to assess the processes of carbon removal, nitrification and denitrification.

In a new approach the wastewater treatment can be based on particular conditions, maintained in the biological reactor, which allow simultaneous nitrification and denitrification, without alternating anoxic and aerobic phases in time or in space. We suggest controlling the aeration system by means of measuring the NO in the off-gas in combination with DO and/or ORP levels. We postulate, that the DO concentration in the aeration tank can be optimised when the aeration is adjusted resulting in the production of NO. The optimal setpoint for the NO emission rate needs to be investigated for every plant. As NO is built during denitrification only, NO-levels in the off-gas exceeding background concentrations indicate that nitrification and simultaneous denitrification takes place. The ORP needs to be adjusted for the specific waste water, but should not be too low; preferably at levels about 100–125 mV. To make sure that the carbon is digested properly TVOC shall be kept low. CO_2 and TVOC help to monitor but also control carbon removal.

There is a possibility of high removal efficiencies for TOC and nitrogen in activated sludge plants which are not equipped with specific denitrification steps. This combination of treatment will also decrease operating costs due to the low DO required in the biological reactor and resulting low energy consumption. For analysis of the effectiveness of such processes further investigations are needed.

An optimal combination of online parameters like ORP, TVOC and NO are effective for controlling the plant and allowing for simultaneous nitrification and denitrification. To find the optimal setpoints for these parameters and to investigate kinetics on NH_4-N, NO_2-N and NO_3-N removal under the discussed conditions, more research is required.

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