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Sulfur Dioxide Treatment from Flue Gases Using a Biotrickling Filter−Bioreactor System

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Complete treatment of sulfur dioxide (SO_2) from flue gases in a two-stage process consisting of a biotrickling filter followed by biological post-treatment unit was investigated. The biotrickling filter could remove 100% of influent SO_2 from simulated flue gas at an empty bed residence time of 6 s for a concentration range of 300−1000 ppm_v. All the absorbed SO_2 was recovered in the biotrickling filter liquid effluent as sulfite (a product of chemical reaction of SO_2) and sulfate (product of biological oxidation of sulfite). The biotrickling filter liquid effluent was further processed biologically in a single post-treatment unit consisting of a combined anaerobic and microaerophilic reactor for the simultaneous reduction of sulfate and sulfite to sulfide and oxidation of sulfide to elemental sulfur. The post-treatment unit could effectively treat the biotrickling filter effluent and produce elemental sulfur. The sulfur production efficiency of the reactor reached about 80% of the SO_2 treated. This new biological treatment system seems to be a promising alternative for flue gas desulfurization.

Introduction

Sulfur dioxide (SO_2) and sulfur oxides (SOx) are some of the main pollutants in many industrial, chemical, and petrochemical processes off-gases. As a result, sulfur oxides have become major atmospheric pollutants, particularly in urban areas. SO_2 has been suspected in several air pollution disasters, notably Donora (U.S.A), the Meuse Valley (Belgium), and several episodes in London (*1*). All fuels used by humans such as coal, oil, natural gas, peat, wood, and other organic matters contain sulfur which will be released as sulfur oxides during combustion. Out of total emissions of SO_2, industrial sectors contributed 69%, mainly from power stations (*2*). SO_2 emissions cause acid rain and adversely affect human health, livestock, and plants.

Various methods exist to reduce SO_2 emissions. The most commonly used ones are the reduction of fuel sulfur through raw material processing or fuel change, increase of stack height and dispersion of source location through proper planning and zoning of industrial areas, and the reduction of pollutant discharge at the source using control equipment such as flue gas desulfurization (FGD) systems. FGD is probably the most widely used technique to control SO_2 emissions from industries (*3*). FGD includes a wide range of physical and chemical methods which can be quite efficient, but FGD often generates additional wastes or requires additional chemical and energy inputs. Clearly, there is a need for environmentally friendly alternatives. Biofiltration, or the use of microorganisms to treat air streams, seems to be one of the most promising alternatives to conventional air pollution control techniques (*4*). Interestingly, the use of biological reactors for the treatment of sulfur oxides has not been greatly explored.

One approach that was considered consists of absorbing sulfur dioxide either in water or in aqueous slurries of limestone and converting it to sulfite and sulfate in a chemical scrubber (see, e.g., ref. *5*). Sulfite and sulfate thus formed can then be reduced under anaerobic conditions to sulfide by sulfate reducing bacteria. In a third reactor, the sulfide can partially oxidized usually by *Thiobacillus* spp. to elemental sulfur under microaerophilic conditions. A disadvantage of this approach is that sulfate reduction to sulfide and subsequent oxidation to sulfur are performed in two separate bioreactors which increases capital costs (*6*). Another approach is to pass the flue gas through a biotrickling filter with *Desulfovibrio desulfuricans*. Under optimum conditions, this organism has been shown to reduce sulfur dioxide to hydrogen sulfide within 1−2 s contact times (*7*). The hydrogen sulfide thus produced is further converted to sulfate. A drawback of this approach is that *D. desulfuricans* is a strict anaerobe, and maintaining anaerobic conditions in biotrickling filters treating flue gases containing on average 2−8% residual oxygen remains a challenge (*8*). Further, this approach requires treating H_2S in a large air stream, and it will produce a dilute sulfate solution of which discharge may be regulated. Hence, the application of this last approach may be limited. Clearly, for optimum SO_2 biotreatment, it is desirable to have a simple system, which combines some of the chemical and biological reactions to produce sulfur rather than sulfate.

In the present study, the development of a new integrated biological system for the complete treatment of SO_2 from flue gases is described. The treatment system consists of a biotrickling filter followed by a single biological post-treatment unit. The proof of concept was demonstrated at the bench-scale, and the performance of the system was monitored under selected operating conditions.

Materials and Methods

Biotrickling Filter Setup and Operation. A schematic of the experimental setup is shown in Figure 1. The reactor was made out of clear Schedule 40 PVC pipe, fittings and caps (i.d. = 4 cm, Ryan Herco, Burbank, CA). The total length of the reactor was 60 cm and the bed height was 50 cm. The reactor contained 0.63 L of packing made of open pore polyurethane foam cubes (4 × 4 × 4 cm, specific surface area of 600 m^2/m^3; density of about 35 kg/m^3 (*9*)) cut to cylindrical shape to fit the reactor internal diameter. The trickling liquid was sprinkled over the packed bed at a rate of 0.8 m/h (1 L/h) from the top of the reactor. A relatively low trickling rate was selected to minimize possible mass transfer limitations toward the attached biofilm. The low trickling rate also minimizes the liquid consumption as the liquid is not recycled as is usually done in biotrickling filters. The biotrickling filter effluent was collected from the bottom of the reactor and fed to the post-treatment unit. The gas inlet and outlets ports were located at the bottom and top lids of the reactor, respectively.

The biotrickling filter was operated at room temperature (20−24 °C). The pH inside the reactor was maintained at 6.8

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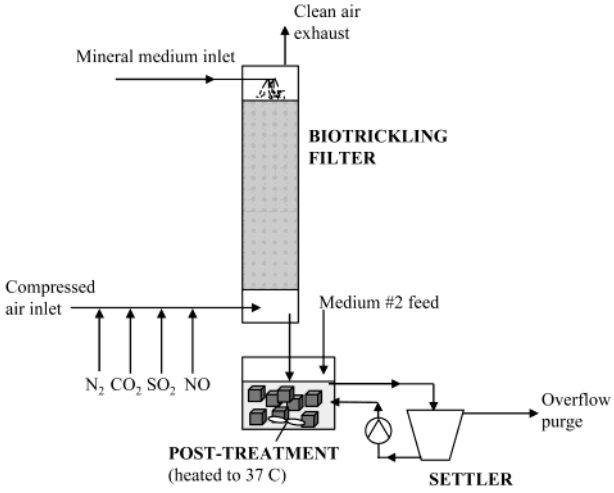

FIGURE 1. Schematic of the experimental setup.

TABLE 1. Summary of Main Operating Events

day	event/conditions
1−31	biotrickling filter was operated at an EBRT of 60 s; gas composition, 75% air, 25% CO_2 and 300−1000 ppm_v each NO and SO_2
31−44	gas composition changed to flue gas composition (75% N_2, 15% air, 10% CO_2, 200−1000 ppm_v each NO and SO_2); EBRT = 60 s
44−51	EBRT was reduced to 30 s
48−80	glucose supply (SO_4−S:COD about 1:1.5) started in the post- treatment unit; post-treatment unit temperature increased to 37 °C and unit is sealed to prevent air entry
51−75	EBRT reduced to 10 s
75−80	EBRT reduced to 6 s
76−80	air supply (as per stoichiometric requirement) in the post-treatment unit

± 0.2 by adding sodium carbonate (0.75 g/L) to the liquid being trickled. The liquid trickled consisted of a mineral medium with the following composition (in g/L in demineralized water) K_2HPO_4 (1); KH_2PO_4 (1); $MgCl_2$ (0.25); $CaCl_2$ (0.52); Na_2CO_3 (0.75) and trace metal solution 1 mL/L (*10*). No nitrate or sulfate was added to the mineral medium as the inlet gas to the biotrickling filter contained NO and SO_2 in an attempt to develop cotreatment of both combustion pollutants. The reactor was seeded with about 200 mL of activated sludge collected from an Orange County Sanitary District sewage treatment plant (Fountain Valley, CA) and with bacteria collected from agricultural soil. There was no external carbon source supply to the biotrickling filter except for CO_2. A timetable of the specific operating conditions is presented in Table 1.

Simulated flue gas was prepared by mixing a metered flow of 15% compressed air, 10% CO_2, 75% N_2, and NO and SO_2 gases (5% each in N_2, Scott Specialty Gases, San Bernardino, CA) according to the required composition. The total gas flow rate was varied to achieve empty bed residence times (EBRT) in the reactor ranging from 6 to 60 s.

Post-Treatment Unit. The post-treatment unit was made out of clear Schedule 40 PVC pipe (ID = 15 cm, Ryan Herco, Burbank, CA) fitted with gastight caps. A schematic of the unit is shown in Figure 1. The total height of the treatment unit was 15 cm with a working volume of 1.7 L. A glucose solution (20 g/L in deionized water) was supplied to the system at a flow rate of 500 mL/day which corresponds to a S−SO_4^{2-}:COD ratio of 1:1.5. Glucose was used as the carbon source for sulfate reducing bacteria in this study, although an inexpensive carbon source would clearly be necessary for industrial application. Glucose, acetate, and lactate were found by others to be comparable carbon sources for sulfate reducing bacteria (*11*). Gentle mixing of the reactor contents was achieved by using a magnetic stirrer. The post-treatment reactor was maintained at a temperature of 35 ± 2 ° C using a hot-stir plate. The pH of the system was maintained at 7−8 by adding Na_2CO_3 whenever required. To improve the biomass holding capacity in the reactor, 10 cubes of polyurethane foam (see above for characteristics) were added to the reactor. A metered flow of air (1 mL/min), calculated from the required half moles of oxygen per mole of sulfate to be oxidized to elemental sulfur, was supplied to the reactor by a peristaltic pump.

Batch Studies for the Determination of the Kinetics of Sulfate Formation. These investigations were carried out to determine the role of microorganism in the biotrickling filter. They were conducted in 250 mL Pyrex bottles (microcosms) fitted with Teflon lined silicon septa. The reaction mixture consisted of 100 mL of mineral medium (see above) and 600 mg_{dw}/L of biomass taken from the biotrickling filter. The microcosms were flushed with nitrogen gas and were then closed immediately. CO_2 and air were added to the microcosms to make the gas composition similar to that of the biotrickling filter inlet. Finally, 10 mL of gaseous SO_2 (100%) was added to each microcosms. The microcosms were incubated at 300 rpm on a rotary shaker (New Brunswick Scientific, U.S.A.). Liquid samples were withdrawn at regular intervals and analyzed for sulfate and sulfite. Control experiments were carried out in the same way except that the reaction mixture was not containing any biomass.

Analyses. The analysis of SO_2 was carried out using a combustion gas analyzer (IMR-1400 Gas analyzer, IMR Environmental Equipment International Inc., U.S.A.). Selected grab samples were analyzed using SO_2 Draeger tubes (Fisher Scientific, U.S.A.). The lower detection limit for each method was 1 ppm_v. Sulfate/sulfite and sulfide analyses were carried out as per standard methods (*12*). Sulfate analysis was based on a spectrophotometrical method. Sulfite was determined by titration using standard potassium iodide-iodate titrant and a starch indicator, with careful precautions to avoid any interferences. Elemental sulfur analysis was done as described by Schedel and Truper (*13*, *14*), by reacting the elemental sulfur with cyanide to produce thiocyanate which was quantified spectrophotometrically as $Fe(SCN)_6^{3-}$. Dissolved oxygen in the liquid phase was measured using polarographic electrode (Thermo Orion, Beverly, MA). pH measurements were carried out using a regular pH meter (Fisher Scientific, U.S.A.).

Results and Discussion

Performance of the Biotrickling Filter. For the initial 30 days, the inlet gas composition was somewhat different from that of real flue gases. The gas stream treated consisted of SO_2 and NO in 75% air and 25% carbon dioxide. Sulfur dioxide and nitric oxide inlet concentrations were in the range of 300−1000 ppm_v each. Under these conditions, at an empty bed residence time (EBRT) of 60 s, the biotrickling filter exhibited over 97% SO_2 removal efficiency irrespective of the SO_2 inlet concentration tested. After this adaptation phase, treatment of simulated flue gases similar to flue gases from coal burning power plants was initiated. The biotrickling filter was initially operated at an EBRT of 60 s; next the EBRT was gradually reduced to 30 s, 10 s, and finally to 6 s (see Table 1). Throughout these investigations, the mineral medium trickling rate was kept constant. The results for SO_2 are shown in Figure 2. In all the cases, the biotrickling filter could remove all influent sulfur dioxide to below the detection limit (1 ppm_v) once it reached steady state.

As far as NO removal is concerned, the original intent was to attempt simultaneous removal together with SO_2. As

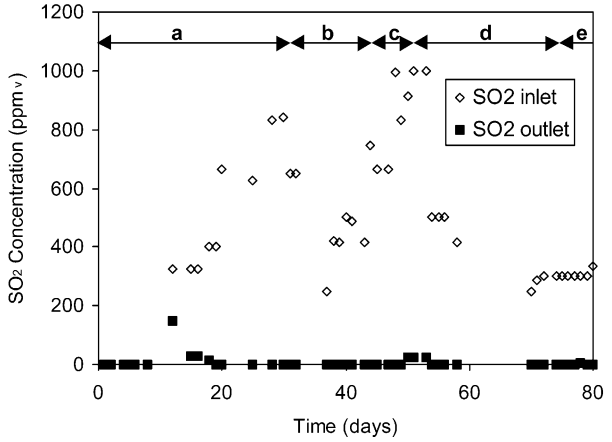

FIGURE 2. Inlet and outlet concentrations of SO_2 in the biotrickling filter. Conditions: (a) startup phase at an EBRT of 60 s, SO_2 in 75% air and 25% CO_2; thereafter operated with simulated flue gas (see Table 1 and text for details) and (b) EBRT of 60 s; (c) 30 s; (d) 10 s; (e) 6 s. SO_2 detection limit was 1 ppm_v.

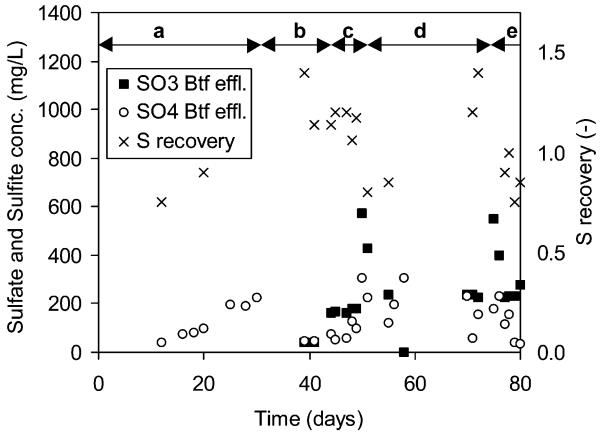

FIGURE 3. Sulfite and sulfate concentrations in the biotrickling filter effluent and recovery of the S−SO_2 removed as S-sulfate and S-sulfite. See Figure 2 caption for the descriptions of the different phases.

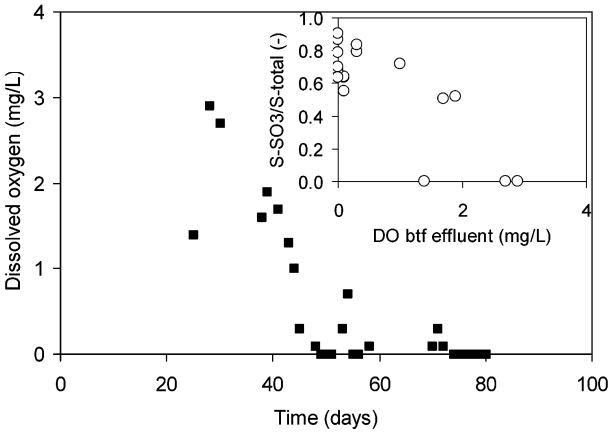

FIGURE 4. Dissolved oxygen of the biotrickling filter effluent vs time. The inset shows the ratio of the S-sulfite to total sulfur recovered in the biotrickling filter effluent as a function of the effluent dissolved oxygen. Note that day 31 marked the beginning of operation at reduce oxygen content in the gas undergoing treatment.

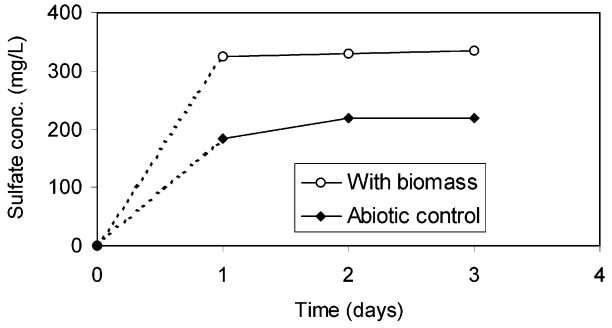

FIGURE 5. Kinetics of sulfate production from SO_2 in microcosms in the presence and absence of biomass (abiotic control is mineral medium only).

reviewed by Stepanov and Korpela (*15*), NO can undergo several biological transformation depending on the conditions. In our case, there were two hypothetical mechanisms that we saw could result in NO treatment. NO could possibly be absorbed in the trickling liquid and be converted chemically to nitrite or biologically to nitrate under aerobic conditions. Under this scenario the resulting nitrite/nitrate containing effluent would be denitrified in the post-treatment unit. The other hypothetical treatment mechanism involved chemolithoautotrophic organisms such as *Thiobacillus denitrificans* (*7*, *16*) in the biotrickling filter which would reduce NO to nitrogen gas under anoxic conditions, while oxidizing sulfite (from absorbed SO_2) to sulfate. Such a reaction had been demonstrated for the oxidation of sulfide (*16*, *17*) which is energetically favorable but has not been proven for the oxidation of sulfite.

Unfortunately, due to analytical problems with NO_x measurements, only qualitative results on the fate of NO were obtained. The data showed that gaseous NO was partially removed; however, great uncertainty exists concerning the exact removal efficiency (removal was about 5−20%). Trace concentrations of nitrite (0.2 −3.8 ppm) were detected in the biotrickling filter liquid effluent, while nitrate concentrations were essentially zero. This corresponds to about 1−10% of the nitrogen load to the biotrickling filter and indicates that some absorption of NO occurred. However because of the large excess of nitrogen gas, a complete nitrogen balance was not possible. Under these circumstances and the apparent failure to achieve reasonable NO removal, the focus of the study was directed toward SO_2 removal.

Sulfate, sulfite, and sulfide concentrations in the outlet of the biotrickling filter were monitored continuously to allow closure of the sulfur balance. As no sulfate or sulfite was added to the mineral medium, inlet concentrations were always zero. As expected, sulfur dioxide reacted with the trickling water to form sulfite. In the presence of oxygen, sulfite is converted to sulfate both chemically and biologically. As shown in Figure 3, the sulfite and sulfate concentrations in the biotrickling filter effluent varied significantly with the reactor operating conditions and with time. As the EBRT was reduced and a biofilm was established, the trend was toward higher sulfite concentration and lower dissolved oxygen concentrations (Figures 3 and 4). At an EBRT of 60 s, almost all sulfur dioxide was converted to sulfate, while at lower EBRTs, sulfite was the dominant product. At an EBRT of 6 s, 90% sulfur dioxide was recovered as sulfite and oxygen became the rate-limiting factor (inset Figure 4). In most cases, the sulfur balance was closed (±20%).

Biological conversion of sulfite to sulfate probably played a significant role in the biotrickling filter as shown from microcosms experiments conducted with cells sampled from the biotrickling filter. The results presented in Figure 5 reveal that both the rate and the yield of sulfate formation were much higher in the presence of microorganisms than in the abiotic controls. Determining the exact contribution of the

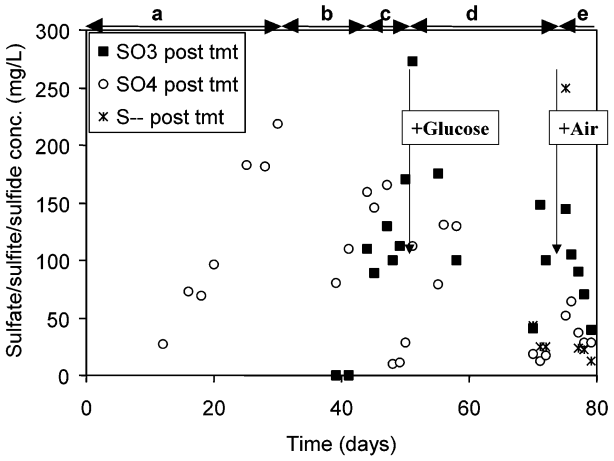

FIGURE 6. Sulfate, sulfite, and dissolved sulfide concentration profiles in the post-treatment unit. Note that scattering is due to the dependency on the reactor conditions (see Figure 2 for the inlet SO_2 concentration and EBRT).

biological processes to the formation of sulfate in the biotrickling filter would require abiotic biotrickling filters to be operated. This was not included in the experimental plan.

Performance of the Post-Treatment Unit. One of the objectives of the present study was to develop a complete biological treatment system using only one biotrickling filter and one post-treatment unit. Therefore, the effluent from the biotrickling filter was fed to a single post-treatment system to be optimized for the recovery of elemental sulfur. The bioreactor combined an anaerobic sulfate/sulfite reduction step and a partial sulfide oxidation step under microaerophilic conditions to produce elemental sulfur. To provide the necessary electron donor for the reduction of sulfate and sulfite to sulfide, glucose was supplied to the system. The $S-SO_4^{2-}$:COD ratio was always kept in the range of 1:1.5 which was the reported optimum for sulfide production (*18*). At a higher COD supply, methanogens are expected to compete with sulfate reducing bacteria. Oxygen was supplied to the system at the stoichiometric requirement for sulfur production from sulfide, i.e., 0.5 mole of oxygen per mole of sulfide.

The concentrations of sulfate, sulfite, sulfide, and hydrogen sulfide in the post-treatment were monitored continuously (Figure 5). For elemental sulfur, it was difficult to determine elemental sulfur formation quantitatively because of the heterogeneity of the system. Part of the sulfur may have been trapped in the foam cubes, in the post-treatment unit. The effect of varying the glucose feed or the oxygen supply was not systematically investigated. However, occasional breakdown of the system resulting in aerobic conditions in the post-treatment unit correlated with low sulfur production and high sulfate discharge indicating that the desired treatment mechanisms were indeed occurring.

At the initial stages of reactor operation, more attention was paid to establish an efficient sulfate reducing process culture. Therefore, during this period no oxygen was supplied. When sulfide production reached around 80% of the total sulfate/sulfite entering the system, oxygen supply was gradually started. The results are shown in Figures 6 and 7. Initially, the sulfide production rate was very low, possibly because of suboptimum conditions or because the inoculum might not have been rich with sulfate reducing bacteria (SRBs). SRBs are strict anaerobes and many have their optimum temperature for growth at 35−37 °C. Hence, on day 48, all reactor ports were carefully sealed to avoid air entry, and the post-treatment temperature was increased to 37 °C. Note that for industrial application, maintaining the post-treatment at about 35−37 °C should not pose any problems as biotrickling filter effluent should be in the order of 50−60 ° C (*19*) and waste process heat should be widely available. After these interventions, sulfide production gradually started and reached a steady state within a week. At this point, a sulfur mass balance over the post-treatment unit revealed that sulfide production was about 80% of the total sulfur entering the system.

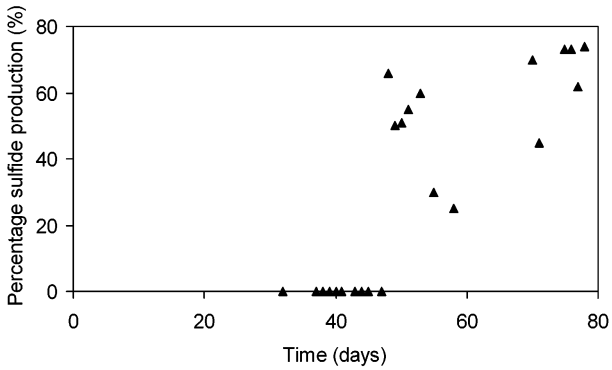

FIGURE 7. Sulfide production in the post-treatment unit.

TABLE 2. Sulfur Production in the Post-Treatment Unit

day	% sulfur produced	day	% sulfur produced
77	43	79	60
78	54	80	80

When sulfide production stabilized to 80% of the sulfate/sulfite entering the post-treatment unit, oxygen supply to the post-treatment unit was started, and elemental sulfur production was observed within 24 h. This rapid response is probably due to the fact that the bioreactor was seeded with acclimatized sulfide oxidizing bacteria enriched and grown on thiosulfate medium (*6*). Moreover, the biotrickling filter effluent contained sulfide-oxidizing bacteria. This was proven by batch experiments (see Figure 5) where microcosms inoculated with effluent of the biotrickling filter gave higher concentration of sulfate compared to abiotic controls. Sulfur production in the post-treatment unit was stable and could be sustained. This proves that anaerobic and microaerophilic zones as described by Okabe et al. (*20*) for differentiated sulfate/sulfite reduction and for sulfide oxidation, respectively, were successfully established.

The amount of elemental sulfur produced was calculated using the mass balance for entire sulfur species present in the system (Table 2) and compared to elemental sulfur analysis. Within 3 days of operation 60−80% of the total sulfur entering in the post-treatment system was converted to elemental sulfur, while the sulfide concentration in the liquid effluent remained below 3 ppm. The remaining sulfur was present as sulfite or sulfate in the effluent. The sulfite/sulfate concentration leaving the post-treatment was in the range of 6−20 mg S/L (Figure 6) which is negligible compared to the total loading of the system. The residual sulfate/sulfite may be due to an excess (either globally or locally) of oxygen in the system. Although the oxygen was supplied in stoichiometric ratio, metering of such a low airflow rate is difficult. Another possible explanation is that some sulfite was converted to sulfate and sulfide by disproportionation (*19*). Further research is needed to optimize oxygen delivery and the yield and rate of sulfur production.

Overall, the SO_2 treatment system that was investigated proved to be robust and effective over a wide range of operating conditions. At this time, a key question is this of the suitability of the process to operate at a higher temperature than tested so far, as combustion gases are expected to be cooled to about 60 °C for coal fired boilers and to about

70 or 80 °C for incinerators. Higher temperatures will obviously result in a decrease of the rate of SO_2 absorption in the biotrickling filter. If needed, cooling of the flue gas may be accomplished using evaporative cooling after dilution of the flue gas with fresh air. But such cooling may not be necessary (or even wanted), as thermophilic sulfate reduction (*21*, *22*) and sulfide oxidation (*23*, *24*) have both been widely reported, which suggest that the main biological processes occurring in the post-treatment are likely to work at higher temperatures. Ultimately, the choice of the process temperature will depend on an integrated design procedure, considering the sizing of the biotrickling filter and of the post-treatment unit together. With the strengthening of environmental legislation and the pressure for environmentally friendly end-of-pipe treatment processes, it appears that the system described herein is a promising alternative for the treatment of SO_2 from flue gases.

Acknowledgments

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