Operational Parameters Analysis for the Removal of H_2S and NH_3 under Transient Conditions by a Biofiltration System of Compost Beds

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Wastewater treatment plants (WWTP) are considered an important source of offensive odors, which are caused mainly by hydrogen sulfide, ammonia, and volatile organic compounds. For the elimination of these pollutants, biofiltration offers an economical alternative of high efficiency and low waste generation. The main factors that affect the performance of a biofilter are the characteristics of the packing material, pH, moisture, concentration of pollutants, and residence time of the gas in the bed. These parameters must be optimized to improve the performance of the biofilter. The purpose of this work was to evaluate the removal efficiency of H_2S and NH_3 with biofilters packed with compost mixtures made from chicken manure and lignocellulosic residues (pruning waste, sugarcane bagasse, and rice husk) under different residence time (60, 45, 33, 25, and 18 s at 40% moisture) and at two levels of pollutant concentration (52 mg H_2S/m^3 and 2 mg NH_3/m^3 and high: 260 mg H_2S/m^3 and 10 mg NH_3/m^3). At 60 s, all biofilters had high removal efficiency (higher than 90%) at the two evaluated gas concentrations. The maximum elimination capacity was reached at 25 s, with a removal efficiency of 80% for both gases at the highest concentration. Also, the effect of moisture content over removal efficiency was evaluated at 25 s. When moisture was 30%, the biofilter of compost from manure and pruning waste decreased its efficiency below 80%; on the other hand, the biofilters with the compost of manure and the two other lignocellulosic residues (sugarcane bagasse and rice husk) reduced their efficiency considerably when the moisture content was 20%. The results showed that the highest removal efficiency was obtained at a residence time of 25 s and a bed moisture content of 40% for the three types of compost mixtures evaluated. These results suggest that the moisture content of the packing material has a significant influence on the elimination of contaminants that are soluble in water. These results are valuable to scale up this kind of biotechnologies in real applications.

1. Introduction

The management and treatment of wastewater are considered significant sources of gaseous emissions, including offensive odors, that cause negative effects on the life quality of the surrounding population. The main compounds that produce odor in a wastewater treatment plant (WWTP) are volatile compounds of sulfur such as hydrogen sulfide (H_2S), nitrogen compounds such as ammonia (NH_3), in addition to volatile organic compounds (VOCs) (Easter et al. 2005). There are a variety of technologies to reduce these emissions and to comply with air quality regulations; biotechnologies, for example, require low capital and operating costs, and are environmentally friendly compared to physicochemical technologies. Among the available biotechnologies, biofiltration is the most used in industries due to its ease of implementation, maintenance, and low cost. In this type of biotechnology, the gases pass through a wet porous bed and are diffused into the aqueous phase of a biofilm containing microorganisms for the degradation of contaminants, as well as nutrients and oxygen (Delhoménie and Heitz 2005).

The main factors that affect the performance of a biofilter are packing material, moisture content, and empty bed residence time (EBRT). The following characteristics of packing material are desirable to obtain a satisfactory removal of gas: high specific surface area (300-1000 m^2/m^3); high porosity (0.4-0.9), optimal water retention capacity to maintain the moisture content between 40-70%, the presence and intrinsic availability of nutrients, and presence of a large and diverse microbial community (Rene et al. 2013). The use of compost as a biofilter bed is frequent because it meets most of the desirable characteristics and, its natural origin makes it usually affordable (Delhoménie and Heitz 2005). The mix of lignocellulosic material with compost helps to prevent rapid degradation and loss of compost structure that drives to a decrease in biofiltration performance (Forero et al. 2018). On the other hand, moisture content of the bed affects the microbial activity of the biofilter and the gas adsorption in the bed, and it should be between 40-70%. At industrial conditions, the air stream may dry the bed, therefore, diminishing moisture content and decreasing the efficiency of the elimination (Delhoménie and Heitz 2005). Then, the moisture content is a critical factor during the industrial process when water is a limited resource or when moisture control is difficult. Finally, empty bed residence time (EBRT), represents the time it takes for the gas to pass through the biofilter; thus, this residence time depends on the bed volume and gas flow rate. The EBRT affects the removal efficiency since the residence time must be enough for the transfer and degradation of the pollutants. This situation usually occurs at a low flow rate or high EBRT; however, these conditions imply a high-volume system, which is a common situation in biofilters used for the removal of volatile compounds mixtures or with concentrated gases (Rene et al. 2013). EBRT depends on the characteristics of the bed material, the nature of the pollutants (hydrophobicity and biodegradability), and their concentration (Delhoménie and Heitz 2005).

These factors must be optimized to achieve the best possible performance of the biofilter. For this reason, the objective of this work was to evaluate the removal of hydrogen sulfide and ammonia at several EBRT and moisture content of bed, in biofilters packed with compost made of chicken manure and mixtures of lignocellulosic materials (pruning waste-CP, rice husk-CA, and sugarcane bagasse-CB). These mixtures previously showed high efficiency and the capacity to remove higher loads of gases (Vela-Aparicio et al. 2020).

2. Materials and Methods

2.1. Biofiltration system

The packing materials were obtained previously from the composting of one lignocellulosic material in a 1:1 volume ratio with two-year-old chicken manure. The chicken manure was collected from a poultry farm located in Nemocón, Colombia. Three different mixtures were obtained: rice husk + manure (CA), pruning waste + manure (CP), and sugarcane bagasse + manure (CB) (Vela-Aparicio et al. 2020). The characteristics of each compost mixture are shown in Table 1.

Table 1: Characteristics of the three mixtures of compost and lignocellulosic materials used as packing bed

Characteristics	pruning waste + manure (CP)	sugarcane bagasse + manure (CB)	rice husk + manure (CA)
Density, g/L	850.45 ± 2.66	753.30 ± 1.53	796.66 ± 0.58
% Size particle distribution,			
> 4.75 mm	77.32	46.60	24.40
4.75- 2.36 mm	2.08	16.56	19.02
2.36 - 1 mm	10.30	9.74	21.64
1 - 0.6 mm	0.78	8.15	11.51
0.65 - 0.25 mm	0.73	6.83	9.15
< 0.25 mm	0.81	3.51	8.20
Water holding capacity (WHC), gH_2O/g material	1.02	1.20	1.68
Buffer capacity, mol H^+/kg material	0.24	0.37	2.80

The biofiltration system in a laboratory scale (Figure 1) consisted of two biofilters per packing material, which were constructed in PVC pipes with a diameter of 10.16 cm, a height of 81 cm, and a total volume of 0.007 m^3 (6.6 L). An ascending gaseous stream through the biofilters was applied using an air compressor (150 pounds). H_2S was volatilized from an acidic solution of Na_2S and NH_3 was volatilized from a solution of NH_4OH 1%, both using the air discharged by a vacuum pump. Afterwards, these gases were mixed with humidified air in a mixing chamber. This mixed stream was finally distributed into the biofiltration system. The

bed moisture content was determined in a thermobalance (PCE- MB-120C) and was adjusted with water when necessary.
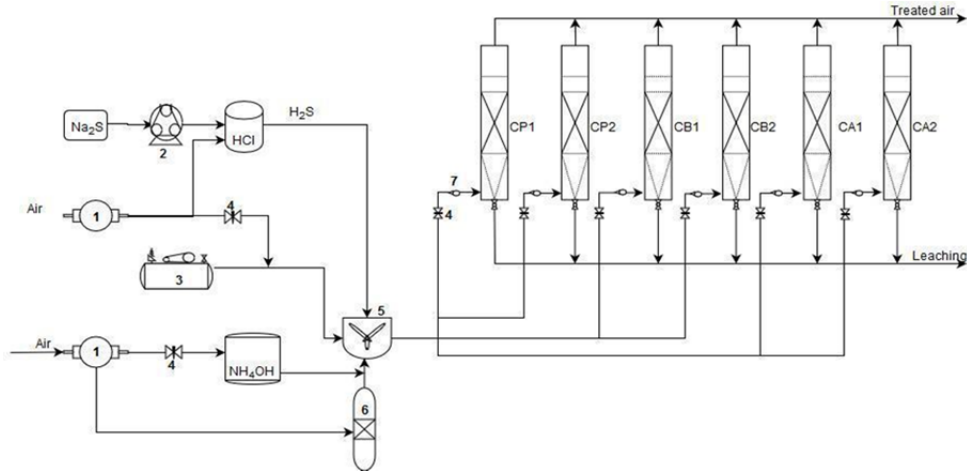

Figure 1: Gas generation system and laboratory-scale biofilters. 1. Vacuum pump; 2. Peristaltic pump; 3. Air compressor; 4. Valve; 5. Mixing chamber; 6. Humidifier; 7. Flowmeters; CP. Pruning waste + manure, CB. Sugarcane bagasse + manure, CA. Rice husk + manure (numbers 1 or 2 indicate replicate biofilter system).

2.2. Gas analysis

The measurements of H_2S and NH_3 were defined and monitored at the inlet and outlet of the biofilters with a portable multi-gas monitor MultiRAE (PGM-6228 RAE Systems) when H_2S concentrations were lower than 100 ppm; a portable detector with a higher H_2S range (Biogas 5000) was used for higher concentrations. The measurements were taken once the gas concentration was steady. The gas sampling was made three times per day, obtaining a daily average of gas removal data. The removal efficiency percentage (%RE) was calculated for each gas using the following equation:

$$\%RE = \frac{(C_i - C_o) * 100}{C_i} \tag{1}$$

where C_i: gas input concentration (mg/m^3), and C_o: gas output concentration (mg/m^3).

The maximum elimination capacity (EC$_{max}$) was calculated at the gas highest concentration using Eq. (2):

$$EC(\frac{g}{m^3h}) = \frac{Q_{gas} * (C_i - C_o)}{V_{bed}} \tag{2}$$

where Q_{gas}: contaminated air flow rate (m^3/h), C_i: gas inlet concentration (g/m^3), C_o: gas outlet concentration (g/m^3), and V_{bed}: bed volume (m^3).

2.3. Evaluation of operational parameters

The gas inlet flow rate (Table 2) was modified through the adjustment of valves installed in the biofiltration system and verified with flowmeters to evaluate the empty bed residence time (EBRT).
Each EBRT was evaluated at two levels of gas concentration: low (52 mg H_2S/m^3 and 2 mg NH_3/m^3) and high (260 mg H_2S/m^3 and 10 mg NH_3/m^3). These concentrations were established as a result of previous monitoring in the pre-treatment zone of the WWTP El Salitre, Bogotá, Colombia (Vela-Aparicio et al. 2019).
As gas inlet load depend on flow rate and gas concentration (Eq. 3), the inlet load increased when EBRT was reduced i.e., flow rate was increased.

$$Gas\ inlet\ load(\frac{g}{m^3h}) = \frac{Q_{gas} * C_i}{V_{bed}} \tag{3}$$

where Q_{gas}: contaminated air flow rate (m^3/h), C_i: gas inlet concentration (g/m^3), and V_{bed}: bed volume (m^3).

The EBRT with the highest elimination capacity was selected to evaluate the effect of moisture content on the performance of the biofilter. The bed was dried with a dry air stream; then, the moisture content was determined in the thermobalance and it was adjusted by adding water to obtain the desired value. The moisture content evaluated was 40%, 30%, 25%, and finally decreased to 20% (dry weight). The evaluation was made at a constant concentration of gases of 75 mg H_2S/m^3 and 3,6 mg NH_3/m^3, which corresponded to

loads between 10-11 g/m^{3}h of H$_2$S and 0.5-0.6 g/m^{3}h of NH$_3$. These concentrations corresponded to the average emissions of H$_2$S and NH$_3$ found during the monitoring (mentioned above) during the dry season.

Table 2: H$_2$S and NH$_3$ load during the evaluation of empty bed residence time (EBRT) at two concentrations

Day	EBRT, s	Flow rate, m^3/h	H$_2$S inlet load, g/m^{3}h	NH$_3$ inlet load, g/m^{3}h
1-5	60	0.39	3	0.1
6-9			16	0.6
10-12	45	0.53	4.2	0.2
13-16			20.5	0.8
16-19	33	0.72	5.7	0.2
19-24			28.5	1.1
24-27	25	0.95	7.5	0.3
28-41			37.3-44.3	1.4
42-48	18	1.31	10.3	0.4
49-50			52.2	2.0
58-62	25	0.95	7.5	0.3
63-65			37.3	1.4

3. Results and Discussion

3.1. Evaluation of EBRT

The experiments were carried out to identify the optimum EBRT and moisture content for the elimination of H$_2$S and NH$_3$ by biofilters made of compost and lignocellulosic material. In the evaluation of EBRT, the removal efficiency of H$_2$S was close to 100% at 60 and 45 s for all biofilters, even at the high level of concentration (Figure 2a). When the EBRT was decreased to 33 s on day 16, the biofilters %RE slightly diminished to 90%. Biofilter CP was the most affected for the high concentration during the evaluation of the residence time of 25 s. This biofilter had a reduction in its %RE at the lower concentration, while in the other biofilters the %RE was higher than 80%. However, the increase of H$_2$S concentration at 25 s caused a drastic reduction of %RE in the three biofilters. A stop period in the gas inlet after the evaluation of 25 s, allowed the biofilters to recover so the efficiency rose again to 80% at EBRT of 18 s and a low concentration level. Nevertheless, when the load was increased to 52 gH$_2$S/m^{3}h, the removal efficiency decreased to below 50%, indicating that the biofilters reached their maximum elimination capacity. The posterior re-evaluation of removal efficiency at 25 s on day 61 showed a slight increase in the %RE of H$_2$S for the three biofilters. This increase could happen because the stop time allows the recovery of microbial activity after microbial stress due to the earlier high H$_2$S load. Cabrol et al. (2016) observed this behavior in the biofiltration of NH$_3$ and VOCs under transient conditions.
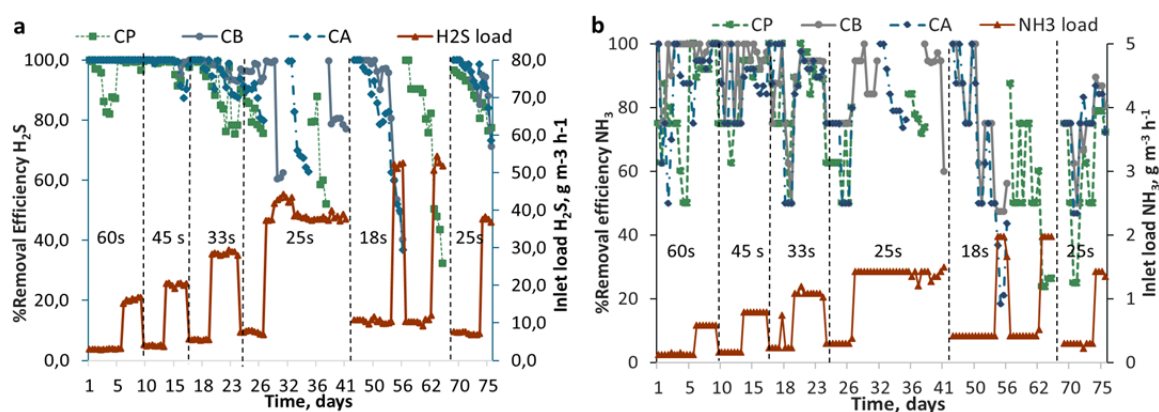

Figure 2. Inlet gas load and removal efficiency of (a) H$_2$S and (b) NH$_3$ in biofilters with different compost mixtures. CA, manure + rice husk; CP, manure + pruning waste; and CB, manure + sugarcane bagasse. Numbers inside correspond to EBRT.

The removal of NH$_3$ showed a similar behavior (Fig 2b) with a reduction of %RE when EBRT was decreased. However, the changes in the concentration resulted in the variability of the removal efficiency. This result could be a consequence of the inhibition of nitrification caused by the high H$_2$S loads, a phenomenon

described previously by Malhautier et al. (2003). Although a removal efficiency higher than 90% for biofiltration of H_2S and NH_3 has been reported at an EBRT of 10 s in a biofilter packed with cured compost (Hou et al. 2016), in this study, similar results were not achieved. The decrease in %RE at low EBRT was possibly due to the accumulation of sulfur and $(NH_4)_2SO_4$, which decreases the superficial area in the bed and reduces the mass transfer of pollutants in the gas phase to the liquid phase (Kim et al. 2002; Tsang et al. 2015).

According to the results shown in Table 3, the biofilter CP had the lowest removal capacity of H_2S and NH_3. This result could be because almost 80% of this material had a particle size higher than 4.75 mm, so there would be a smaller area for biofilm formation and, therefore, for the transfer and oxidation of gas in the bed. This situation was reported previously for similar packing material (compost from municipal solid waste and pruning residues) in the biofiltration of composting gases. (López et al. 2011).

Table 3: Maximum elimination capacity (EC_{max}) of each biofilter packing bed. These values were reached at an EBRT of 25 s

Packing bed	EC_{max} H_2S, g /m^3h	EC_{max} NH_3, g /m^3h
CP	29,3	1,1
CB	32,8	1,2
CA	32,0	1,2

Finally, an EBRT of 25 s was selected to evaluate the effect of moisture content in the biofiltration of H_2S and NH_3 because the maximum elimination capacity of biofilters was reached in this time and the removal efficiency for both gases was higher than 80% for the highest concentration of gases. For the application in an industrial system, low residence times of the gas are desirable because it would allow treating high loads of the pollutants. Also, the biofilters should maintain a high %RE in transient conditions such as shock loads, as the one applied during this evaluation since these conditions are common in industrial systems.

3.2. Evaluation of moisture content

At a moisture content of 40%, the removal efficiency for both gases in biofilters CA and CB was above 90%, while in biofilter CP was up to 65% (Fig.3). When moisture was reduced to 30% (days 7 to 10), the removal efficiency of NH_3 decreased slightly, but the %RE for H_2S diminished by almost 15%. As the moisture content was decreased to 20%, the %RE for H_2S diminished to less than 40% in biofilters CB and CP. In contrast, %RE in biofilter CA was 75%. This difference could occur due to the physical characteristics of each material, like the water holding capacity, which is related to the retentivity, i.e., how easily water is retained in the packing material independently of the moisture content (Dorado et al. 2010). Since the water holding capacity is higher for the CA bed than in the other two packing materials (Table 1), this material could maintain for a longer time the water in the biofilter and avoid a drastic decrease in the microbial activity.
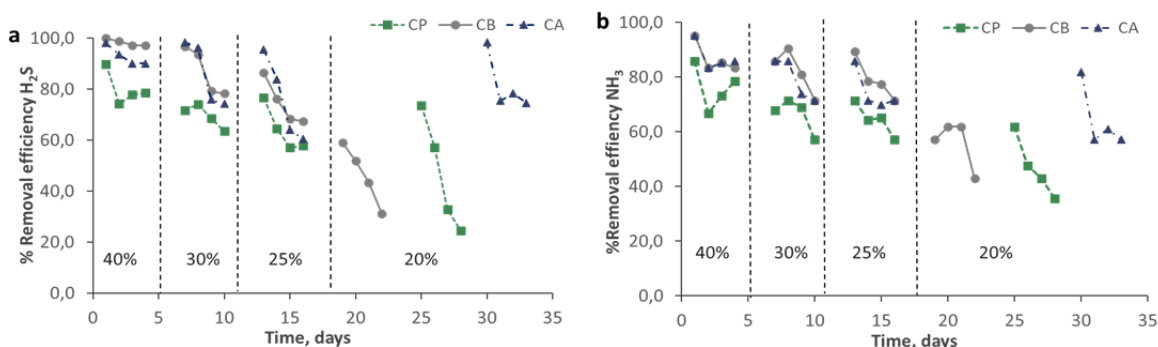

Figure 3. Moisture content and removal efficiency of (a) H_2S and (b) NH_3 in biofilters with different compost mixtures; CA, manure + rice husk; CP, manure + pruning waste; and CB, manure + sugarcane bagasse. Numbers inside correspond to moisture content.

The reduction in the removal efficiency as the moisture decreases could occur due to the loss of the microbial biofilm present on the surface of the bed particles, thus, affecting the adsorption, transfer, and degradation of the gases. This reduction could also be caused by the formation of preferential channels in the beds because the gas distribution is not homogeneous and causes a decrease in the contact area with the biofilm (Rene et al. 2013). According to these results, 40% is the optimum moisture content for the biofiltration of H_2S and NH_3 with the compost mixtures used. These packing materials have adequate water availability and optimal aeration for the physicochemical process and microbial activity.

4. Conclusions

For the biofiltration of gas pollutants, the residence time of the gas and moisture content of the bed are critical parameters to obtain high performance. In the elimination of H_2S and NH_3 by biofilters with packing bed of compost mixture of lignocellulosic wastes and chicken manure, EBRT of 25 s allows high removal efficiency and the highest elimination capacity for both gases. In the evaluation of moisture content, 40% was suitable for the elimination of this type of contaminants by the evaluated compost biofilters. Finally, compost mixture of sugarcane bagasse and chicken manure showed the highest elimination capacity with high removal efficiency (87% H_2S and 86% NH_3) for the elimination of H_2S and NH_3 under transient conditions. Thus, this packing bed is recommended for additional studies to evaluate its use at an industrial level in the elimination of offensive odors in WWTP and other industries, such as solid waste management and composting.

Acknowledgments

References

Cabrol L, Poly F, Malhautier L, Pommier T, Lerondelle C, Verstraete W, Lepeuple AS, Fanlo JL, Roux X Le (2016) Management of Microbial Communities through Transient Disturbances Enhances the Functional Resilience of Nitrifying Gas-Biofilters to Future Disturbances. Environ Sci Technol 50:338–348 . doi: 10.1021/acs.est.5b02740

Delhoménie M-C, Heitz M (2005) Biofiltration of air: a review. Crit Rev Biotechnol 25:53–72 . doi: 10.1080/07388550590935814

Dorado AD, Lafuente FJ, Gabriel D, Gamisans X (2010) A comparative study based on physical characteristics of suitable packing materials in biofiltration. Environ Technol 31:193–204 . doi: 10.1080/09593330903426687

Easter C, Quigley C, Burrowes P, Witherspoon J, Apgar D (2005) Odor and air emissions control using biotechnology for both collection and wastewater treatment systems. Chem Eng J 113:93–104 . doi: 10.1016/j.cej.2005.04.007

Forero DF, Acevedo P, Cabeza IO, Peña C, Hernandez M (2018) Biofiltration of acetic acid vapours using filtering bed compost from poultry manure - pruning residues - rice husks. Chem Eng Trans 64:511–516 . doi: 10.3303/CET1864086

Hou J, Li M, Xia T, Hao Y, Ding J (2016) Simultaneous removal of ammonia and hydrogen sulfide gases using biofilter media from the biodehydration stage and curing stage of composting. Environ Sci Pollut Res 23:20628–20636 . doi: 10.1007/s11356-016-7238-4

Kim HS, Kim YJ, Chung JS, Xie Q (2002) Long-term operation of a biofilter for simultaneous removal of H_2S and NH_3. J Air Waste Manage Assoc 52:1389–1398 . doi: 10.1080/10473289.2002.10470871

López R, Cabeza IO, Giráldez I, Díaz MJ (2011) Biofiltration of composting gases using different municipal solid waste-pruning residue composts : Monitoring by using an electronic nose. Bioresour Technol. 102:7984–7993 . doi: 10.1016/j.biortech.2011.05.085

Malhautier L, Gracian C, Roux JC, Fanlo JL, Le Cloirec P (2003) Biological treatment process of air loaded with an ammonia and hydrogen sulfide mixture. Chemosphere 50:145–153 . doi: 10.1016/S0045-6535(02)00395-8

Rene ER, Kennes C, Veiga MC (2013) Biofilters. In: Kennes C, Veiga MC (eds) Air Pollution Prevention and Control: Bioreactors and Bioenergy, First. John Wiley & Sons, Ltd., pp 60–72

Tsang YF, Wang L, Chua H (2015) Simultaneous hydrogen sulphide and ammonia removal in a biotrickling filter: Crossed inhibitory effects among selected pollutants and microbial community change. Chem Eng J 281:389–396 . doi: 10.1016/j.cej.2015.06.107

Vela-Aparicio DG, Muñoz Lasso CC, Brandão PF, Cabeza IO, Hernandez MA (2019) Evaluation of H_2S, NH_3 and Volatile Organic Compounds Emissions in a Waste-Water Treatment Plant in Bogota, Colombia. In: Air Pollution Conference Brazil | 4th CMAS South America. Belo Horizonte, Minas Gerais, Brazil, p Session 10

Vela-Aparicio D, Forero DF, Hernández MA, Brandão PFB, Cabeza IO (2020) Simultaneous biofiltration of H_2S and NH_3 using compost mixtures from lignocellulosic waste and chicken manure as packing material. Environ Sci Pollut Res. *In press.* doi: 10.1007/s11356-020-10817-w