

Reviewer 2

Reviewer comments and suggestions	Responses from authors
The authors mainly focus on problems caused by high sulfur content in the used substrates/feedstock. They should give some examples for such substrates and evaluate their relevance. In practice this problem is of minor importance, because biowaste/biosolids (which are the main substrates for small-scale AD) consist of carbohydrates, lipids and proteins.	The entire manuscript has been re-written to accommodate more of chemistry features that did not include the issue of sulfur in the substrate since this is of a minor importance in anaerobic digestion (AD). These chemistry features were perceived as
Carbohydrates and lipids don't contain sulfur (only C, H and O). Only proteins can be a source for sulfur if they contain one or both amino acids cysteine and/or methionine.	This is true and as previously mentioned the issue of sulfur in AD is minor. As such, it has been totally removed from the manuscript. Instead, other issues such as accumulation of VFAs and how to avoid them in order to improve process efficiency have been discussed.
In practice other problems like inhibition caused by high nitrogen content or accumulation of VFAs (especially acetic acid) caused by inappropriate feeding (too high volumetric load) leading to pH drop are of much greater importance.	As previously mentioned, the entire manuscript has been re-written with more chemistry features and how important these features are in process optimization. Methods to avoid the accumulation of VFAs were also identified and discussed from a chemistry standpoint.
Page 9, lines 292 – 294: The authors state that the effects of sulfur and bacterial interaction within the AD process have not been adequately explained. There are many publications about the bacterial interaction and the role of sulfur in the AD process and possible inhibitions by sulfur (especially in connection with modelling and simulation of the biological processes and extensions/modifications of the used models like the well-established ADM 1). A short selection of publications is	Because the manuscript has been re-written, the line number pointed out by the reviewer now has new information. As such, information about the effects of sulfur no longer appears anywhere in the manuscript. The short selection of publications listed by the reviewer was carefully perused and it was noted that information about sulfur inhibition in AD processes is totally exhaustive hence the issue of sulfur was removed in our review.

listed below:

- Kalyuzhnyi, S. and Fedorovich, V. (1998) Mathematical modelling of competition between sulphate reduction and methanogenesis in anaerobic reactors. *Biores. Tech.* 65, 227-242.
- Fedorovich, V., Lens, P. & Kalyuzhnyi, S. (2003). Extension of Anaerobic Digestion Model No. 1 with processes of sulfate reduction. *Applied Biochemistry and Biotechnology*, 109(1-3), 33-45.
- Xavier Flores-Alsina, Kimberly Solon, Christian Kazadi Mbamba, Stephan Tait, Krist V. Gernaey, Ulf Jeppsson, Damien J. Batstone, Modelling phosphorus (P), sulfur (S) and iron (Fe) interactions for dynamic simulations of anaerobic digestion processes. *Water Research*, Volume 95, 15 May 2016, Pages 370-382.
- E.L. Barrera, H. Spanjers, K. Solon, Y. Amerlinck, I. Nopens, J. Dewulf, Modeling the anaerobic digestion of cane-molasses vinasse: Extension of the Anaerobic Digestion Model No. 1 (ADM1) with sulfate reduction for a very high strength and sulfate rich wastewater. *Water Research*, 71 (2015), pp. 42-54
- Ernesto L. Barrera, Henri Spanjers, Jo Dewulf, Osvaldo Romero and Elena Rosa. The sulfur chain in biogas production from sulfate-rich

liquid substrates: a review on dynamic modeling with vinasse as model substrate. J Chem Technol Biotechnol, 2013, 88, 1405-1420 • H.-H. Chou, J.-S. Huang, W.-G. Chen, R. Ohara, Competitive reaction kinetics of sulfate-reducing bacteria and methanogenic bacteria in anaerobic filters. Bioresource Technology, 99 (17) (2008), pp. 8061-8067 • T. Hao, P. Xiang, H.R. Mackey, K. Chi, H. Lu, H. Chui, M.C.M. van Loosdrecht, G.-H. Chen. A review of biological sulfate conversions in wastewater treatment. Water Research, 65 (2014), pp. 1-21 • J.B.M. Klok, M. de Graaff, P.L.F. van den Bosch, N.C. Boelee, K.J. Keesman, A.J.H. Janssen. A physiologically based kinetic model for bacterial sulfide oxidation. Water Research, 47 (2) (2013), pp. 483-492 • H������ Hauduc, Bruce Johnson, Charles Bott, Matthew Ward, Imre Tak������. Incorporating Sulfur and Relevant Reactions into a General Plantwide and Sewer Model. IFAC PapersOnLine 50-1 (2017) 3935��3940	
Page 13, lines 322 and 323: The authors conclude that the disappearance of hydrogen and the accumulation of VFAs are linked to inhibitory effects of sulfur. These interactions should be described in more detail.	This part of the manuscript, now on page 14, lines 391 to 395 has been clarified. The inhibitory effects of sulfur were no longer accommodated in our clarification; instead, the reasons that could lead to hydrogen disappearance and accumulation of VFAs in the digestion process were dwelt on.

Page 13 and Figure 1 on page 14: First the authors describe the effects of sulfur and H₂S, but the following explanations and the reaction scheme in Figure 1 are dealing with the inhibition effects caused by halogenated aliphatic hydrocarbons (HAHs)! It has to be explained (and verified by literature) if this reaction scheme can really be transferred to the inhibition effects caused by sulfur.	The figure in question (Figure 1) has been removed as the figure and its information does not correspond with the present content of the manuscript given the fact that over 90% of the manuscript has been re-written.
Page 16, chapter 7.1: The authors suggest to use analytical methods (like NDIR) to determine the exact amount of sulfur and the usage of techniques for sulfur removal. These suggestions don't seem to be reasonable, because sophisticated analytical methods like NDIR are normally not used in small-scale biogas plants (especially not in developing countries). A small-scale biogas plant for developing countries has to be cheap, easy to use and with the lowest technical expenditure. Techniques for sulfur removal are normally not used in practice (not even in highly industrialised countries), because this would be too expensive. In addition, other opportunities are used to prevent problems caused by too high sulfur content. The optimum dosage of substrates (and optimum mixture of different substrates) can be calculated by simulation using an appropriate model (e.g. extended ADM 1 resp. ADM 1xp with incorporated processes for sulfur removal and inhibition).	The issue of sulfur no longer appears anywhere in the manuscript. As a result, the suggested methods of using analytical techniques such as NIR for sulfur evaluation have also been removed from the manuscript. In fact, this section now appears on page 18 and 19 as section 6, 6.1, and 6.2 respectively. This was due to the major changes made to the manuscript and specifically described methods to accelerate and optimize AD through the use of non-biological conductive materials with the ability to absorb toxic compounds and stimulate interspecies hydrogen transfer (IHT) and hydrogen partial pressures (HPP) via combining AD with microbial electrochemical systems (MES).
Page 16, chapter 7.2: The authors suggest the combination of small-scale AD with a microbial electrochemical system. How could this be put into practice (in which way could MES be	In page 16, section 7.2, now page 18 section 6.2, the methods by which AD can be integrated with a MES is briefly described.

implemented into a digester)?	
Page 17, line 497: The authors mention the use of magnetic fly ash for pretreatment of mainly livestock manure. This has to be described in more detail. What is the effect of magnetic fly ash (supply with trace elements? precipitation of S2-?...)?	This has been described in more detail and highlighted in line 652 to 655 on page 21, section 7. The trace elements have also been provided.
Summary of specific comments	Responses from authors
Furthermore, there are a number of minor errors that should be corrected. The text contains a number of mistakes and missing blank spaces (e. g. between value and unit):	The entire manuscript has been spell-checked to avoid minor mistakes such as those pointed out by the reviewer.
Page 1 (Introduction) line 42: The last sentence should be partitioned "...from system's digestate. Small scale AD systems..." instead of "...from system's digestate, small scale AD systems ...".	This part has been deleted due to major changes made in the manuscript.
Page 2 (Table 1) "Remarks": "The spread of pathogens is also prevented." instead of "The spread of pathogens are also prevented."	This has been noted and corrected accordingly.
Page 3 (Table 1) "Advantages": "biosolids" instead of "bio solids".	Noted and corrected.
Page 3 (Introduction) line 61: The expression "...must be nifty..." is inappropriate and should be reworded.	This part has been totally erased from the manuscript due to major changes made.
Page 4 (2.1.1. Hydrolysis) line 86: The expression "...chemistry standpoint..." is inappropriate and should be reworded (e.g. "chemical perspective" or "chemical point of view").	This expression has been reworded to suit reviewer's choice of words.

Page 4 (2.1.1. Hydrolysis) line 91: The chemical equation (1) is incorrect, because it doesn't describe a cleavage of polymers (in this form it seems to be a kind of addition!). $C_6H_{10}O_5$ cannot be used as a molecular formula for carbohydrates, it should be $(C_6H_{10}O_5)_n$. In addition, the hydrolysis of carbohydrates doesn't produce elemental hydrogen(!). Also there is a missing blank space between "2" and "H₂O" ("2 H₂O" instead of "2H₂O").	The chemical equation (1) is noted and corrected as well as the missing blank space between "2" and "H₂O". However, we disagree with the reviewer's comment on hydrolysis of carbohydrates that it does not produce elemental hydrogen. It should be noted that in most cases, biomass including those used as feedstock in AD is made up of large organic polymers. For microorganisms in the digestion process to access the energy potential of the biomass, polymer chains must first be broken down into smaller constituent parts known as monomer units in the form of simple sugars, amino acids and fatty acids, given off hydrogen. These compounds are directly used by the microorganisms in the digestion process. The process of breaking these chains and dissolving the smaller molecules into solution is called hydrolysis hence it is regarded as the necessary first step in AD. In fact, equation (1) is construed to mean: Biomass + H₂O → monomers + H₂. The following articles will help the reviewer refresh his memory on the reaction stages of AD: • P. Bajpai (2017). Anaerobic Technology in Pulp and Paper Industry, SpringerBriefs in Applied Sciences and Technology, DOI 10.1007/978-981-10-4130-3_2. • K.C. Surendra, Devin Takara, Andrew G. Hashimoto, Samir Kumar Khanal (2014). Biogas as a sustainable energy source for developing countries: Opportunities and challenges. Renewable and Sustainable Energy Reviews 31, 846–859. • https://en.wikipedia.org/wiki/Anaerobic_digestion. • https://www.e-education.psu.edu/egee439/node/727.
Page 4 (2.1.1. Hydrolysis) lines 105 and 106: The expression "the hydrolysis stage" is repeated within the same sentence ("...in the hydrolysis stage;..." can be deleted). Also this sentence should be partitioned "...to soluble organic compounds. The microorganisms responsible for..." instead of "...to soluble organic compounds in the hydrolysis stage; the microorganisms responsible for..."	This has been noted, corrected and partitioned as suggested by the reviewer.

Page 4 (2.1.1. Hydrolysis) lines 106 – 108: The sentence “..., so that organics that are insoluble in H ₂ O are solubilized by the H ₂ O to render chemical bonds broken for the formation of soluble compounds that can be used by bacteria cells [28-31].” is not comprehensible and should be reworded.	This part of the manuscript, which now appears in line 166 to 169, has been rephrased to make better sense.
Page 4 (2.1.2. Acidogenesis) line 116: “Whereas the production of volatile fatty acids (VFAs) is increased...” instead of “Whereas the production of volatile fatty acids (VFAs) are increased...”	This sentence now appears in line 177. It has been corrected accordingly.
Page 4 (2.1.2. Acidogenesis) lines 120 – 122: Missing blank spaces between number and molecular formula (e.g. “2 H ₂ O” instead of “2H ₂ O”).	This has been adjusted accordingly. Blank spaces have been added between number and molecular formula. The equations now appear in lines 181 to 185.
Page 5 (2.1.3. Acetogenesis) line 135: Misleading wording (“...hydrogen-scavenging bacteria that can be converted to CH ₄ [23].” The whole bacteria will not be converted to CH ₄ !	This line, now line 199, explains how the hydrogen produced from the AD process is used up by methane-producing microorganisms.
Page 5 (2.1.3. Acetogenesis) lines 137 – 139: Missing blank spaces between number and molecular formula (e.g. “2 H ₂ O” instead of “2H ₂ O”).	The lines, 137 to 139 are now 202 to 206. The missing blank spaces between number and molecular formula have been added.
Page 5 (2.1.3. Acetogenesis) line 137: “HCO ₃ -“ instead of “HCO ₃ –“.	This now appear in line 202 and have been corrected.
Page 5 (2.1.3. Acetogenesis) lines 141 and 142: wrong molecular formula for acetate (“CH ₃ COO-“ instead of “CHOOH”).	This is now in line 210 and has been corrected.
Page 5 (2.1.3. Acetogenesis) line 149: “acetate” instead of “acetates”.	This now appears in line 217 and has also been corrected.
Page 5 (2.1.4. Methanogenesis) line 151: “2.1.4. Methanogenesis” instead	This now appear in line 219 and have been corrected.

of “2.1.4. Acetogenesis”.	
Page 5 (2.1.4. Methanogenesis) lines 165 and 166/Page 6, line 167: Missing blank spaces between number and molecular formula (e.g. “2 H ₂ O” instead of “2H ₂ O”).	The blank spaces were noted in lines 166 and 167, which are now lines 234 and 235 respectively. However, blank spaces have been added.
Page 7 (2.1.4. Methanogenesis) line 213: The expression “...that feed the AD process.” should be reworded.	The statement now appears in lines 285 and 286 on page 9. It has been rephrased for better understanding.
Page 8 (3. Current status of small scale AD systems in developing countries) line 255: “etc.” instead of “etc”.	This section of the manuscript has been totally removed due to changes made.
Page 8 (3. Current status of small scale AD systems in developing countries) line 256: “...over 40 million small scale AD systems exist...” instead of “...over 40 million small scale AD systems exists...”.	This section has been removed from the manuscript.
Page 8 (3. Current status of small scale AD systems in developing countries) lines 257 and 263: “m3” instead of “m3”.	Section 3 has been removed from the manuscript.
Page 12 (4. Overview of previous research on AD) Table 4: “...wastewater treatment plants...” instead of “...wastewater treatment plant...”.	This now appear on page 14 and has been corrected.
Page 12 (4. Overview of previous research on AD) Table 4: “...1 : 4...” instead of “...1:4...”.	This has been corrected. Blank space was added as “1 : 4”.
Page 13 (5. Issues addressed in small scale AD processes) lines 335 and 336: The expression “changing needs” is not comprehensible and should be reworded.	This section is now section 4 and now on page 15. The expression “changing needs” has been deleted.

Page 13 (5. Issues addressed in small scale AD processes) line 340: The expression "...for ambiguity avoidance sake." is not comprehensible and should be reworded.	Page 13 is now page 15, and line 340 now lines 421 and 422. The expressions on these lines have been rephrased.
Page 14 (6. Existing key technical barriers and research knowledge gaps) line 379: "The flow of electrons is also channelled..." instead of "The flow of electrons are also channelled..."	The section has been reworded and the expression on line entirely removed.
Page 15 (6. Existing key technical barriers and research knowledge gaps) line 392: "There seems to be an obligate,..." instead of "There seem to be an obligate,..."	Line 392 changed to 429 and the expression has been corrected.
Page 15 (6. Existing key technical barriers and research knowledge gaps) line 407: The expression "...must be nifty..." is inappropriate and should be reworded.	The expression, now on page 16 and in line 443, has been reworded. The word 'nifty' has been replaced by the word 'dexterous'.
Page 15 (6. Existing key technical barriers and research knowledge gaps) line 408: The expression "hitches" is inappropriate and should be reworded (e.g. "problems" or "difficulties").	This expression now appears on page 16 and the word 'hitches' has been replaced with 'difficulties'.
Page 15 (6. Existing key technical barriers and research knowledge gaps) line 414: "...and hydrogen depends on these..." instead of "...and hydrogen depend on these..."	This was noted and corrected on page 16 in line 450.
Page 15 (6. Existing key technical barriers and research knowledge gaps) line 408: The expression "digester environment" is inappropriate and should be reworded or explained.	The expression 'digester environment' could not be located in line 408. Perhaps, it has been removed, since the manuscript underwent major changes.

Page 15 (6. Existing key technical barriers and research knowledge gaps) lines 420 and 421: “Since this discovery, no clear explanation has been provided how this influences AD processes.” instead of “Since this discovery however, no clear explanation have been provided as to how this influences AD processes.”.	This has been corrected accordingly in lines 455 and 456 on page 16.
Page 16 (7. Potential solutions to address key technical barriers) line 455: The sentence should be partitioned “...increases process efficiency. Knowledge of feedstock...” instead of “...increases process efficiency; knowledge of feedstock...”.	The major changes made to the manuscript have affected this expression as it has been deleted to accommodate other information relevant to the content of the revised sections of the manuscript.
Page 18 (8. Recent advances in the small scale AD technology) line 506: “...advanced AD technology,...” instead of “...advance AD technology,...”.	This expression has been reworded and now appears on page 21 in line 658. Now section 7.
Page 18 (8. Recent advances in the small scale AD technology) line 507: “...to convert the organic fraction...” instead of “...to convert organic fraction...”.	This has been corrected on page 21 in lines 659, precisely now section 7.
Page 18 (9. Future directions and opportunities) line 523: “...where there seems to be...” instead of “...where there seem to be...”	This expression has been totally erased from the manuscript due to changes made.
Page 18 (9. Future directions and opportunities) line 524: The phrase “...,reliable plants that can be locally manufactured” does not match with the first part of the sentence and should be reworded.	This expression has also been deleted.
Page 18 (9. Future directions and opportunities) line 532: “...and offer the opportunity...” instead of “...and	This expression has also been erased from the manuscript.

offers the opportunity...”.	
Page 18 (9. Future directions and opportunities) line 533: “...and provide...” instead of “...and provides...”.	The expression has been deleted.
Page 18 (9. Future directions and opportunities) lines 542 and 543: The phrase “...by huge strides in efficiency ease of use,...” is not comprehensible and should be reworded.	This expression can be found in section 8, lines 708 and 709 respectively. The expression has been reworded.
Page 18 (9. Future directions and opportunities) line 547: “farmers” instead of “famers”.	This expression has been erased due to major changes.
Page 20 (References) lines 614 – 616 (9.): “Niles, J.O.; Brown, S.; Pretty, J.; Ball, A.S. & Fay, J.” instead of “Niles J.O., Brown S., Pretty J., Ball A.S., & Fay J.” Page 20 (References) lines 614 – 616 (9.): There are two different years stated (2002 and 2008) for the same publication.	This reference now on page 24, reference no. 6, has been noted and corrected. The correct publication year is 2002.
Page 21 (References) line 656 (27.): “van Haandel, A.; van der Lubbe, J.” instead of “van Haandel A, van der Lubbe J.”	The reference now appears on page 25 and as reference no. 36. It has been corrected.
Page 21 (References) line 659 (28.): “Yadvika, S.; Sreekrishnan, T.R.; Kohli, S.; Rana, V.” instead of “Yadvika Santosh, Sreekrishnan, T.R.; Kohli S.; Rana V.”	This has been noted and corrected. The reference now appears on page 25 and as reference no. 37.
Page 21 (References) line 661 (29.): “Broughton, A.D.” instead of “Broughton A.D.”	This reference also now appears on page 25 and as reference no. 38. It has been corrected accordingly.
Page 22 (References) line 713 (53.): “von Blottnitz, H.” instead of “von Blottnitz H.”	This reference has been totally erased from the manuscript.

Page 24 (References) lines 827 and 828 (101.): “McCarty, P.L. One hundred years of anaerobic treatment. In Anaerobic digestion; Hughes, P.E.; Stafford, D.A.; Wheatley, B.I.; Baader, W.; Lettinga, G.; Nyns, E.J.; Verstraete, W.; Wentworth, R.L.; eds.,” instead of “McCarty P.L. One hundred years of anaerobic treatment. In Anaerobic digestion.; Hughes P.E., Stafford D.A., Wheatley B.I., Baader W., Lettinga G., Nyns E.J., Verstraete W., Wentworth R.L., eds.,”	This reference now appears in reference no. 92 on pages 28 and has been corrected.
Page 25 (References) line 885 (126.): “Parawira, W.” instead of “Parawira W.”	This reference has been erased due to major changes in the manuscript.
Missing blank space between value and unit or/and between values and “–“ (e.g. “70 %” instead of “70%”; “20 – 45 °C” instead of “20–45°C”): Page 5: lines 148, 149.	Blank space added in line 216 of the manuscript.
Page 6: lines 177 – 180, Table 2.	Blank spaces have been added between value and units in lines 249 to 252 including Table 2, and the sections is now 2.2 “The percentage composition of CH ₄ from AD”.
Page 7: Table 3, lines 203, 224, 225.	Blank space has been provided in the first values in Table 3, i.e., 50 – 60 (%) in lines 275 (previously line 203), 297 (previously 224), and 299 (previously 225).
Page 8: lines 226, 276.	Blank spaces have been added in lines 299 (previously 226). However, because of major changes, the information in line 276 has been deleted.
Page 10 and 11: Table 4.	Blank spaces have been provided where needed in Table 4.
Page 17: line 480.	Blank space have also been provided in line 480, now line 628 in section 6.2 on page 20.