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Weiterentwicklung von Verfahren zur Biogasproduktion aus organischen Reststoffen und Energiepflanzen im Zielsystem Umwelt, Energie und Klimaschutz

vorgelegt von

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Inhaltsverzeichnis

Kurzfassung

Der schonende Umgang mit natürlichen Ressourcen und Energie ist eine der wichtigsten Aufgaben der Zukunft. Dies gilt insbesondere für die Bereiche Umweltschutz, Abfallwirtschaft sowie den Ausbau Erneuerbarer Energie und Klimaschutz. Internationale Richtlinien und deren nationale Umsetzung definieren Rahmenbedingungen und ein komplexes Zielsystem, das die Entwicklung innovativer Technologien forciert. Die Anaerobtechnik kann in den genannten Themenfeldern in unterschiedlichen Aufgabengebieten vielfältig eingesetzt werden.

In dieser Arbeit werden kontinuierliche Verbesserungen und die Entwicklung neuer Prozessschritte im Bereich der Biogastechnologie im geltenden Zielsystem diskutiert. In den inkludierten Publikationen werden dazu angewandte Forschungsergebnisse aus Versuchen im Labor- Pilot und großtechnischen Maßstab präsentiert. Verfahrenstechnische Weiterentwicklungen sind sowohl in traditionellen Einsatzfeldern, wie der Behandlung von Abfällen, als auch in neuen Anwendungen, wie bei der Erzeugung von Erneuerbarer Energie aus Energiepflanzen unbedingt notwendig, um die Effizienz der Technologie zu erhöhen und die Kosten zu reduzieren.

In den Versuchen und Berechnungen werden Lösungsansätze zu folgenden zwei Problemstellungen erarbeitet: einerseits zu Prozessketten der Verwertung von organischen Reststoffen und Nebenprodukten mit dem Schwerpunkt auf proteinreichen Fraktionen und andererseits zur Vergärung von Energiepflanzen.

Reststoff- und Nebenproduktverwertung problematischer Substrate

Bei der Verwertung von Reststoffen werden die Vorteile der Co-Vergärung den Herausforderungen und Chancen der Mono-Vergärung von proteinreichen Schlachtabfällen gegenübergestellt.

In Pilotversuchen zur Co-Vergärung von organischen Siedlungsabfällen und tierischen Nebenprodukten aus der Lebensmittelindustrie wurde ein stabiler Abbauprozess bei einer hohen organischen Raumbelastung von 10 $kg_{COD}\,m^3\,d^{-1}$ erreicht, der im Rahmen des Projekts Teil der Entscheidungsgrundlage für die Erweiterung einer Großanlage war. Eine weitere Erhöhung der Raumbelastung auf 12 $kg_{COD}\,m^3\,d^{-1}$ durch die Zugabe von tierischem Fett führte zu einer Akkumulation von organischen Säuren. Die Implementierung einer zweiten Vergärungsstufe zur Verbesserung des Abbaus wurde daraufhin untersucht.

Verfahrensentwicklungen zur Mono Vergärung von tierischen Nebenprodukten wurden am Beispiel einer Anlage, die in einen Schlachthof integriert ist, untersucht. In langjährigem Monitoring und durch intensive Prozesskontrolle konnte trotz hoher Stickstoffkonzentrationen von > 7,5 g kg^{-1} konnte ein stabiler anaerober Abbauprozess implementiert werden. Eine Steigerung der Verwendung von noch zur Verfügung stehenden tierischen Nebenprodukten, vor allem Blut, ist im bestehenden Anlagensystem dauerhaft aber nicht möglich ohne die Stabilität des Abbauprozesses zu gefährden.

Um zusätzliche Nebenproduktströme verwerten zu können, wurde ein Verfahren vorgeschlagen, mit dem das Ammonium direkt aus dem Prozess entfernt werden kann. Die Reduktion des Stickstoffgehaltes in den Fermentern sollte zu einer Verbesserung der Abbauleistung führen. In Vorversuchen wurde der erste Schritt, die Trennung des Ammoniums von den vorliegenden Proteinen durch Hydrolyse und mikrobiologische Desaminierung, untersucht. Zur Entfernung des Ammoniums wurde eine kontinuierliche Pilotanlage zur Ammoniakstrippung getestet, mit der eine ausreichende Entfernung und Rückgewinnung des Ammoniums nachgewiesen werden konnte. Beim vorgeschlagenen Strippverfahren direkt aus dem Prozess ist keine Rezirkulation mit Prozesswasser aus dem Gärrest erforderlich.

Die positiven Auswirkungen der Stickstoffreduktion auf den Vergärungsprozess wurden in kontinuierlichen Versuchen nachgewiesen. Bei einer Reduktion des Gesamtstickstoffgehalts von 7 g kg^{-1} auf 4 g kg^{-1} konnte eine Steigerung der Abbaurate um 40 % nachgewiesen werden. Nahezu der gesamte anaerob verfügbare Kohlenstoff wurde zu Biogas umgesetzt. Bei der Simulation zukünftiger Betriebszustände der Großanlage konnte gezeigt werden, dass nach einer Implementierung des Stickstoffentfernungsprozesses entweder die Raumbelastung durch zusätzliche Zugabe von tierischen Nebenprodukten wie Blut um 60 % gesteigert oder alternativ die Aufenthaltszeit um 25 % reduziert werden kann.

Durch die energetische Verwertung von tierischen Nebenprodukten in standortintegrierten Biogasanlagen und die stoffliche Verwertung des Gärrests als Dünger wird der anthropogene Stickstoffkreislauf geschlossen und damit wichtige Ziele der Abfallwirtschaft und des Umweltschutzes erfüllt.

Biogasproduktion aus Energiepflanzen

Der zweite Schwerpunkt der inkludierten Publikationen ist die Verwertung von Energiepflanzen zu Biogas. Die Gewinnung höchstmöglicher Methanerträge der eingesetzten Substrate unter realen Betriebsbedingungen ist von entscheidender Bedeutung für die Wirtschaftlichkeit der Technologie. An einer Großanlage wurde der Betrieb mit zwei Substratmischungen (100 % Mais sowie 48 % Mais + 52 % Gras/Klee) ohne Güllezugabe über 20 Betriebsmonate die Stoff- und Energieströme bilanziert.

Beim Einsatz von Gras- und Kleesilagen ist durch den höheren Faseranteil, im Vergleich zu Mais Ganzpflanzensilage, eine höhere Flüssigkeitszugabe zur Einstellung des Trockensubstanzgehaltes notwendig, der die Rührbarkeit sicher stellt. Die Prozessflüssigkeit aus dem abgepressten flüssigen Gärrest gewonnen. Durch gesteigerte Separation und Rezirkulation reduzierte sich die Aufenthaltszeit in den Fermentern von jeweils 65 Tagen auf 34 Tage. Die langsamere Verfügbarkeit der Grünlandpflanzen, die reduzierte Aufenthaltszeit und das Abpressen und Ausschleusen des festen Gärrests führten zu einer geringeren Methanausbeute bei den eingesetzten Substraten. 70 % des Restmethanpotenzials wurden in der auf die Masse bezogen mit 7 % kleinen Fraktion des festen Gärrest unter idealen Laborbedingungen gefunden. Um die Auslastung der Anlage sicher zu stellen, müssen an der Großanlage daher um 6 % mehr Substrate zugeführt werden, als im Vergleichsfall der Verwendung von Mais.

Durch das geringere Preisrisiko bei Gras und Klee gegenüber Mais ist der Mehrverbrauch bei der Neuheit der Technologie vertretbar. Zukünftig muss der Ausgärgrad verbessert werden. Die Bilanzierungen legen dar, dass ein maximaler Ertrag durch optimale Führung und intensive Kontrolle des Prozesses erreicht werden kann.

Die Erkenntnisse der durchgeführten Projekte zeigen, dass bei der Weiterentwicklung der Anaerobtechnologie eine noch stärkere Verbindung der Grundlagenforschung zu mikrobiotechnologischen Fragestellungen mit den Herausforderungen an Großanlagen geschaffen werden muss. Die Ausrichtung auf gegebene oder zukünftige Ziele im Umweltschutz, in der Abfallwirtschaft, bei Erneuerbaren Energien oder der Reduktion von Treibhausgasemissionen muss dabei frühzeitig gegeben sein.

Bei der Nutzung von Biomethan als Quelle Erneuerbarer Energie wird es in Zukunft wichtig sein, eine nachfrageorientierte Herangehensweise zu finden. Die Integration in industriellen Anwendungen ist eine bereits etablierte Möglichkeit. Ein weiterer Schritt könnte die Umsetzung von Projekten sein, die Biomethan als alternatives Ökogas vermarkten und die vorhandene Infrastruktur und Anwendungstechnologien für Erdgas nutzen.

Abstract

The sustainable management of natural resources and energy is one of the most important future challenges. This applies in particular for the areas of environmental protection, waste management as well as for the expansion of renewable energies and climate protection. International directives and the national implementation of these define the regulatory framework and a complex system of future objectives, which forces the development of innovative technologies. Anaerobic digestion is a proven technology with great future perspectives in the mentioned field of applications.

In this work, continuous improvements and the development of new process steps in the field of biogas technology are discussed within the given legal and economic environment. The included papers and publications present results from applied research experiments in laboratory, pilot and full scale. Further improvements of processes are absolutely essential in both, traditional applications of anaerobic digestion as the treatment of wastes and by-products as well as in new applications like the production of renewable energy from energy crops. The efficiency of the process chain has to be increased and the treatment costs must be reduced.

Experimental results are focused on two research areas: The first one is, the optimisation of process chains for the degradation of organic wastes, in particular protein rich by-products and the second one focuses on the treatment of energy crops.

Degradation of organic wastes and problematic substrates

The benefits of co-digestion of various waste fractions are compared with the challenges and chances of the mono-digestion of protein rich animal by-products from slaughterhouses.

The co-digestion of the organic fraction of municipal wastes and animal by products from the food industry was tested in pilot scale experiments. A stable degradation process was established at an organic loading rate of 10 $kg_{COD}\ m^3\ d^{-1}$. Within the project this results were part of the decision making for the enlargement of an industrial scale digestion plant. A further increase of the organic loading rate to 12 $kg_{COD}\ m^3\ d^{-1}$ was achieved by adding animal fat and led to an accumulation of organic acids. Thus, the implementation of a second digestion step was tested to increase the degradation performance.

A new process approach has been proposed for the mono-digestion of animal by-products at an anaerobic digestion plant, which is integrated in a slaughterhouse site. At this full scale plant a stable anaerobic degradation could be implemented based on long time monitoring and intensive process control at high nitrogen concentrations of > 7.5 g kg^{-1}. A further increase of the treatment of locally available animal by-products, in particular pig blood, is not possible using the existing plant configuration without the risk of process instabilities and reduced degradation rates.

To accomplish the possibility for the utilisation of additional waste fractions a process is proposed, that enables the removal of ammonia directly out of the digestion process. The reduction of the nitrogen concentration in the fermenter may increase the microbiological degradation performance significantly. The first step of this new process – the separation of ammonia from proteins by hydrolyses and microbiological deamination – was tested in laboratory experiments. A continuous stripping unit was constructed and operated to remove the ammonia from the digestion liquid. A satisfactorily reduction and recovery of ammonia could be achieved. Thus, the complete process chain can be designed smarter, because no dilution by the recirculation of process water is necessary.

The positive consequences of the removal of nitrogen on the digestion process were proved in continuous digestion experiments. The reduction of the nitrogen content from 7 g kg^{-1} to 4g kg^{-1} led to an increase of the degradation rate by 40 % and to a complete conversion of anaerobic available carbon to biogas. In the set of experiments various future operation conditions for the full scale plant were simulated. The results show, that after the implementation of the nitrogen removal process either the organic loading rate could be increased by 60 % or alternatively the retention time can be reduced by 25 %.

The utilisation of animal by-products in biogas plants, which are integrated in the production site, and the recycling of the digestion residues as fertiliser close the human nitrogen cyle. Thus, major goals of the waste management and environmental protection are fulfilled.

Biogas from energy crops

The second main focus of the included papers is the utilisation of energy crops to biogas. The main goal must be to gain the highest possible methane yield under real process conditions. This is essentially important for the economic success for this new application. The material and energy flows have been balanced at a full scale biogas plant for 20 months. In this time the plant has been operated based on two substrate mixes (100 % maize silage and 48 % Maize + 52 % grass-clover-silage) and without any addition of liquid manure.

The higher fibre content of grass and clover compared to the maize plant led to a higher demand for process liquid to dilute the digester content. A dry substance content, that assures the continuous stirring, has to be adjusted all the time. The process liquid is gained by separating the digestion residues in a solid and a liquid phase. An increase in separation and recirculation reduces the retention time from 65 days in each fermentation step to 34 days each. The slower degradation performance of grass and clover, the reduced retention time as well as the separation and discharge of solid digestion residues led to a lower methane yield. 70 % of the residual methane potential was derived in the solid phase, which is 7 % of the total residual mass. The experiments were conducted under ideal laboratory conditions. To assure the full contracted operation of the plant, 6 % more substrate have to be fed in the full scale plant to compensate the losses in residual methane potential.

The reduced price risk of grass and clover compared to maize excuses the additional substrate demand for this new technology application. But, the degradation rate and the methane yield have to be increased in near future developments. The balances show, that an optimal yield can be accomplished by intensive process control.

The results of this work point out, that the future development of anaerobic digestion has to address the connection from basic research on microbiological questions with the challenges of industrial scale plants. An early focus on future goals in environmental protection, waste management, renewable energy and the reduction of green house gas emissions has to be set.

Teil I Rahmenschrift

1. Einleitung

Ein wesentlicher Bestandteil einer nachhaltigen Entwicklung ist der schonende Umgang mit den der Menschheit insgesamt zur Verfügung stehenden natürlichen Ressourcen. Wichtige Bereiche der Nutzung natürlicher Ressourcen durch den Menschen sind insbesondere die Abfallwirtschaft und der Energieverbrauch. In der Abfallwirtschaft wurden in Europa und Österreich in den letzten Jahrzehnten große Fortschritte erreicht. Der Umbau des Energiesystems von fossilen zu Erneuerbaren Quellen beginnt gerade und ist dringend geboten wie die Analysen der Internationalen Energieagentur zeigen. Sie sprechen von einer Energierevolution, die den weltweiten Zugang zu Energie sichern muss und von einer Dekarbonisierung des Energiesystems, was die Abkehr von fossilen Ressourcen bedeutet und in den nächsten 20 – 30 Jahren zu einer weltweiten Reduktion der Treibhausgasemissionen führen soll (IEA, 2008 und IEA, 2009).

Integrierte Technologien, wie die anaerobe Verwertung von organischen Substanzen zu Biogas und Erneuerbare Energie, leisten dazu bereits heute einen wichtigen Beitrag. Die Rolle dieser Technologien wird und muss in den nächsten Jahren noch gestärkt werden. Dazu ist es notwendig, ständige Verbesserungen in allen Schritten der Umwandlungsprozesse zu erreichen und Problemlösungen von der Grundlagenforschung bis zur Optimierung bei industriellen Anlagen anzubieten.

Verbindender Hintergrund bei der Wahl der Anaerobtechnologie zur Verwertung von Reststoffen und bei der Nutzung von nachwachsenden Rohstoffen sind internationale und nationale rechtliche sowie strategische Rahmenbedingungen. Die meisten der vielfältigen Einsatzmöglichkeiten der Anaerobtechnologie sind auf diesen Rahmen zurückzuführen. Die tatsächliche regionale Ausprägung hängt von den zu lösenden Herausforderungen, den zur Verfügung stehenden organischen Ressourcen und den örtlich geltenden Regelungen und Förderbedingungen ab.

3.1. Rahmenbedingungen der Energie-, Klima- und Umweltpolitik

1.1.1. Verwertung von Reststoffen und Nebenprodukten als Ziel in der Abfallwirtschaft

Die Abfallwirtschaft wurde in den letzten Jahrzehnten zu einer wachsenden Aufgabe im Umwelt- und Klimaschutz. Durch Landverbrauch und unkontrollierte flüssige Emissionen in den Boden und in Gewässer sowie Methanemissionen in die Atmosphäre ist die Deponierung von organisch abbaubaren Abfällen heute in Europa keine Option mehr. Die europäische Abfallrahmenrichtlinie 2008/98/EG definiert eine fünfstufige Zielhierarchie, bei der die Vermeidung vor Verwertungsschritten steht und die Beseitigung erst die letzte Stufe darstellt. An einer konkreten Verordnung für organische Abfälle und der damit erhofften Stärkung der anaeroben Verwertung wird noch gearbeitet. 2008 stellte die Europäische Kommission dazu ein Grünbuch vor, in dem der Addition von energetischer Nutzung und dem Schließen von stofflichen Kreisläufen im Sinne der Ressourcenschonung noch nicht die gewünschte Bedeutung beigemessen werden (Europäische Kommission, 2008). Die striktere Trennung und Bereitstellung organischer Reststoffe begünstigt die Umsetzung effizienter Anlagen in einer ausreichenden Ausbaugröße.

Eine besondere Beachtung gilt dem Gefahrenpotenzial tierischer Nebenprodukte (TNP). Die unzureichende Behandlung von Schlachtabfällen oder die unangebrachte Verwendung daraus hergestellter Produkte führte in Europa zu pandemischem Auftreten von Tiererkrankungen wie BSE. Die Verordnung mit Hygienevorschriften für nicht für den menschlichen Verzehr bestimmten tierischen Nebenprodukten 2009/1069/EG (ersetzt 2002/1774/EG) regelt die Verwendungsmöglichkeiten und

Verarbeitungsvorschriften von tierischen Nebenprodukten und teilt die TNP in drei Gefährdungskategorien ein. Auch die Verwertung der Nebenprodukte in Biogasanlagen inklusive der Hygienesierung und des Inverkehrbringens des Gärrests sind geregelt.

1.1.2. Neue Ziele durch Klimaschutz und erneuerbare Energie

Ein neues Zielsystem wurde mit dem Kyoto-Protokoll zur Reduktion der Treibhausgasemissionen gestartet und in den letzten Jahren durch das Energie- und Klimapaket der Europäischen Union auf einen Zeithorizont bis 2020 erweitert. In der Richtlinie 2009/28/EG zur Förderung der Nutzung von Energie aus Erneuerbaren Quellen werden die Ziele für den Ausbau Erneuerbarer Energie in den EU-Mitgliedsstaaten dargelegt. Bis 2020 soll der Anteil Erneuerbarer Energie am Endenergieverbrauch in der EU von 8,5 % auf 20 % gesteigert werden. In einem Zuteilungsverfahren wurde dieses Ziel auf die 27 Mitgliedsstaaten umgelegt. Österreich muss demnach seinen Anteil Erneuerbarer Energie am Bruttoendenergieverbrauch von ca. 24 % auf 34 % in den nächsten 10 Jahren steigern. Ausgehend von vorliegenden Potenzialabschätzungen wurde die Ausrichtung der österreichischen Energiepolitik in einem Prozess und anschließender Erstellung der Energiestrategie Österreich neu definiert (BMWFJ und BMLFUW, 2010). Weitere Ziele definieren die Reduktion der Treibhausgasemissionen um -20 % in Europa. Österreich muss dabei in den Bereichen außerhalb des Emissionshandels, vor allem Raumwärme, Verkehr, Landwirtschaft und Abfallwirtschaft eine Reduktion von -16 % erreichen (Umweltbundesamt, 2010).

3.2. Biogastechnologie als integrierter Beitrag zu Erneuerbarer Energie und Umweltschutz

Biogaserzeugung

Der Abbau von Biomasse unter Sauerstoffausschluss ist ein in der Natur weit verbreiteter Vorgang und von enormer Bedeutung für den Stoffkreislauf der Ökosysteme. Dabei werden hochmolekulare organische Substanzen wie Kohlenhydrate, Lipide und Proteine durch eine anaerobe Mikroorganismenbiozönose letztendlich zu Methan und Kohlendioxid zersetzt. Der anaerobe biologische Abbau von organischen Komponenten erfordert eine Vergesellschaftung verschiedener Bakteriengruppen, die das entsprechende Substrat schrittweise in einem vierstufigen Prozess aus Hydrolyse, Acidogenese, Acetogenese und Methanogenese zu den Endprodukten Methan (CH_4) und Kohlendioxid (CO_2) umsetzen.

Die biochemischen Grundlagen des anaeroben Abbaus organischer Substanzen und die Vorgänge in den einzelnen Phasen des anaeroben Abbaus, der Prozessablauf und zahlreiche Einflussfaktoren und deren Auswirkungen auf den Abbauprozess wurden in Fachbüchern und wissenschaftlichen Publikationen umfassend dargestellt (Braun, 1982; Speece, 1996; Mata-Alvarez, 2002; Ahring, 2003; Bischofsberger et al., 2005; Scholwin et al., 2009). Bei der Darstellung der Herausforderungen und den in den Publikationen dargestellten Lösungsansätzen wird auf die prozesstechnischen konkreten Hintergründe eingegangen.

In Biogasanlagen werden diese Gärungs- und Fäulnisprozesse gesteuert mit den Zielen, die organische Substanz abzubauen, ein energetisch nutzbares Gas zu erzeugen und Nährstoffe in einem Kreislauf zu halten. Anaerobe Abbauwege sind besonders für die Verwertung von heterogen zusammengesetzten oder hoch konzentrierten, feuchten organischen Abfällen und Abwässern, Nebenprodukten oder Energiepflanzen geeignet.

Entwicklung der Technologie

Die Verwertung von organischen Materialien zu Biogas durch anaerobe mikrobiologische Fermentation hat lange Tradition. Geschichtliche Überlieferungen berichten von Faulgruben in Siedlungen, bei denen das entstehende Gas direkt verbrannt und zur Beheizung oder Nahrungszubereitung verwendet wurde.

Die technischen Entwicklungen dieses Vergärungsprozesses in den letzten 150 Jahren sind vielfältig. Sie führen außerdem zu den noch zu lösenden Herausforderungen in der Gegenwart und Zukunft, um die Erwartungen in diese Technologie in Bezug auf Erneuerbare Energie- und Umweltschutzdienstleistungen erfüllen zu können (Baserga et al., 1991; Bischofsberger, 2005). In Tabelle 1 sind die wichtigsten Anwendungsgebiete der Anaerobtechnologie und die bedeutendsten dahinterstehenden Rechtsmaterien für den Bereich des Umweltschutzes und der Abfallwirtschaft sowie den Zielen des Ausbaus Erneuerbarer Energie und des Klimaschutzes aufgelistet.

Bischofsberger (2005) dokumentiert den technischen Einsatz der Anaerobtechnologie am Beginn des Industriezeitalters mit der Behandlung von Siedlungsabwässern und der Stabilisierung von aeroben Klärschlämmen. Hygienische Probleme in stark wachsenden Industriestädten in England und Deutschland brachten die Entwicklung voran. Umweltschutz, die Reinhaltung der Oberflächengewässer und die Verringerung der Seuchengefahr standen dabei im Vordergrund. Das Gas aus den Faultürmen vor Ort wurde für den Wärmebedarf der Kläranlage verwendet.

Mit der Industrialisierung der Landwirtschaft in Westeuropa und der Intensivierung der Schweine- und Rinderhaltung ab etwa 1965 waren große Mengen Gülle an den landwirtschaftlichen Betrieben verfügbar. Zum Schutz der Grundwasserressourcen wurden maximale Nährstoffmengen je Hektar und Ausbringungsverbote in der vegetationsfreien Zeit festgelegt (BMLFUW, 2007b). Aus der Notwendigkeit Gülle zu lagern und zu stabilisieren, Geruchsemissionen zu vermeiden und durch die Möglichkeit der energetischen Nutzung des Gases, entstanden die ersten landwirtschaftlichen Biogasanlagen (Braun, 1986; Baserga et al., 1991).

Tabelle 1: Wichtigste Anwendungszwecke der Biogastechnologie und die dahinterstehenden Rechtsmaterien

Anwendungszweck der Biogastechnologie	Umweltziele und Abfallwirtschaft	Klimaschutz und erneuerbare Energie
Abwasserreinigung und Schlammstabilisierung	EU-Nitratrichtlinie 91/676/EWG Wasserrechtsgesetz 1959 und branchenspezifische Abwasseremissionsverordnungen	
Verwertung von organischen Siedlungsabfällen	Abfallrahmenrichtlinie 2008/98/EG Richtlinie über Abfalldeponien (Landfill directive) 1999/31/EC Grünbuch über die Bewirtschaftung von Bioabfall in der Europäischen Union (Europäische Kommission, 2008) Abfallwirtschaftsgesetz, 2002 Deponieverordnung, 2008 (Nov. 2009)	Abfallrahmenrichtlinie 2008/98/EG Richtlinie über Abfalldeponien (Landfill directive) 1999/31/EC Erneuerbare-Energien-Richtlinie (EG), 2009/28/EG
Verwertung von tierischen Nebenprodukten und anderen Reststoffen aus der Lebensmittelindustrie	Verordnung über tierische Nebenprodukte 2009/1069/EG ersetzt 2002/1774/EG BREF-Doc: Slaughterhouses and Animal By-products Industries, European Commission, 2005 Tiermaterialiengesetz (TMG), 2003 Wasserrechtsgesetz, 1959 und branchenspezifische Abwasseremissionsverordnungen	Erneuerbare-Energien-Richtlinie (EG), 2009/28/EG
Verwertung von Energiepflanzen und landwirtschaftlichen Reststoffen	Österreichisches Aktionsprogramm zum sachgerechten Einsatz von Gärresten zur Umsetzung der EU-Nitratrichtlinie 91/676/EWG Nachhaltigkeitsanforderungen an die Nutzung fester und gasförmiger Biomasse bei Stromerzeugung, Heizung und Kühlung (Europäische Kommission, 2010)	EU Energie und Klimapaket 2020; Europäische Kommission, 2008 Erneuerbare-Energien-Richtlinie (EG), 2009/28/EG Klimastrategie (BMLFUW, 2002) und Anpassung der Klimastrategie, (BMLFUW, 2007a) Energiestrategie Österreich (BMWFJ und BMLFUW, 2010) Ökostromgesetz, 2009

Anaerobe Verwertung von Abfällen und Nebenprodukten

Die Festlegung auf umweltpolitische Zielsetzungen (Vermeidung und Verwertung) in der Abfallwirtschaft in den 1990er Jahren waren ausschlaggebend für den Einsatz der Anaerobtechnologie bei der Behandlung des organischen Anteils von Siedlungsabfällen. Die komplexe Zusammensetzung und die Einbettung in nicht vergärbare Störstoffe machten oft eine umfangreiche Aufbereitung der Abfälle notwendig (Mata-Alvarez, 2002). Bei der Behandlung von industriellen organischen Nebenprodukten aus der Lebensmittel- und Getränkeproduktion beschleunigte die Konzentration auf große Produktionseinheiten die Notwendigkeit die Rest- und Nebenprodukte zu verwerten (Russ und Schnappinger, 2007). Die EU-Verordnung 2002/1774/EG (ersetzt durch 2009/1069/EG) mit Hygienevorschriften für nicht für den menschlichen Verzehr bestimmte tierische Nebenprodukte wurde in Folge der Skandale und Seuchen im Zusammenhang mit Lebensmitteln erlassen. Gemeinsame mit dem Dokument über die besten verfügbaren Technologien für Schlachthöfe und Tierkörperverwertung (BREF document) (European Commission, 2005) öffnen sie somit eine streng geregelte Option, Reststoffe und Nebenprodukte aus Schlachthöfen in Biogasanlagen zu verwerten. Umwelt- und Hygieneziele stehen dabei im Vordergrund. In einem in der Arbeitsgruppe verfassten Buchbeitrag stellen Kirchmayr et al. (2007) [Publikation 4] dieses Thema in Bezug auf die Nutzung tierischer Nebenprodukte in Biogasanlagen umfassend dar.

Der aktuelle Statusbericht Abfallwirtschaft 2009 weist für Österreich 746.000 Tonnen tierische Nebenprodukte aus. Davon stammen 390.000 Tonnen aus der Schlachtung oder der Fleischverarbeitung. Der Rest verteilt sich auf Markt- und Cateringabfälle mit tierischen Anteilen, der Milchproduktion und Falltieren. Der größte Anteil (332.000 Tonnen) wird in Tierkörperverwertungsanlagen entsorgt. Bereits 215.000 Tonnen werden in Biogasanlagen verwertet (Umweltbundesamt, 2009). Im Bundesabfallwirtschaftsplan 2006 wurden für das Jahr 2004 nur 90.000 Tonnen ausgewiesen (BMLFUW, 2006).

Biomethanproduktion für Erneuerbare Energie

Das aktuellste Einsatzgebiet der Biogastechnologie ist die Vergärung von Energiepflanzen zur Erzeugung von Erneuerbarer Energie. Seit dem Inkrafttreten des Erneuerbaren Energie Gesetzes in Deutschland im Jahr 2000 (aktuelle Fassung: EEG, 2009) und dem bundesweiten Ökostromgesetz in Österreich 2002 (aktuelle Fassung Ökostromgesetz, 2009) hat die Erzeugung und Nutzung von Biogas, insbesondere in der Landwirtschaft, erheblich zugenommen. Biogas leistet damit einen Beitrag, die Ziele zur Steigerung der Erneuerbaren Energie und des Klimaschutzes zu erreichen. Als Substrate kommen zunehmend Energiepflanzen, in Mitteleuropa vor allem Mais, zum Einsatz. Die Zielrichtung bei der technischen Planung und der wirtschaftlichen Kalkulation ist seither stärker auf die Optimierung der Energieerzeugung und auf die Erfüllung energiepolitischer Ziele ausgerichtet. Die Integration in das Energiesystem durch das Anbieten von Strom, Wärme und Treibstoff ist eine wichtige Zukunftsaufgabe bei der Weiterentwicklung der Technologie.

Die neue, starke Ausrichtung der Biogastechnologie auf Energieproduktion macht eine Neuinterpretation der bisher im Vordergrund stehenden Umweltziele notwendig. In einigen Anwendungsfällen, wie der intensiven Produktion von Energiepflanzen, insbesondere Mais, stehen diese in teilweiser Konkurrenz zu Umweltzielen (Ifeu, 2008).

Dadurch werden auch die Schwerpunkte in der technischen und ökonomischen Optimierung neu gesetzt: War es zum Beispiel bei der Verwertung organischer Abfälle durch die Entsorgungserlöse ökonomisch sinnvoll unter Einhaltung von Grenzwerten des Umweltschutzes möglichst viele Abfälle zu behandeln, so ist bei der Verwendung von produzierten Energiepflanzen die möglichst hohe Nutzung des Energiegehalts von hoher wirtschaftlicher Bedeutung. Der Anbau von nachwachsenden Rohstoffen ist, anders als die Bereitstellung von Rückständen, Nebenprodukten und Abfällen, mit vergleichsweise hohen Aufwendungen für Dünger und Pflanzenschutz und Bodenbearbeitung verbunden. Damit verbunden kann eine Produktion und Nutzung von Energiepflanzen zur Biogaserzeugung höhere ökologische Belastungen verursachen (Scholwin et al, 2006).

Die Ziele des Umweltschutzes kommen dabei bei der nachhaltigen Produktion der Energiepflanzen und der Ökobilanzierung in einer neuen Form zu tragen.

Die Europäische Kommission berichtet dazu im Juni 2010 an das EU Parlament, wie Nachhaltigkeitskriterien auf Basis der Erneuerbaren Energie Richtlinie (2009/28/EG) in den Mitgliedsländern umgesetzt werden sollen (Europäische Kommission, 2010). Faulstich und Greiff (2008) analysieren, dass die Nutzung der land- und forstwirtschaftlichen Flächen für Nahrungsmittel, Rohstoffe sowie für Energie (Wärme, Strom und Mobilität) zwangsläufig zu Nutzungskonkurrenzen bezüglich der begrenzten Anbaufläche führen muss. Die Produktion der Energiepflanzen muss daher im Sinne der Technologie transparent durch Ökobilanzen und die Reduktion von Treibhausgasemissionen dokumentiert werden.

In der öffentlichen Darstellung wurde bis vor wenigen Jahren die Gewinnung und Nutzung von Biomasse weitgehend als dezentrale Technologie wahrgenommen und galt daher von vornherein als umweltverträglich und nachhaltig. Die industrielle Nutzung und der weltweite Handel mit Biomasse zur Energienutzung machen es notwendig transparente Kriterien einer nachhaltigen Prozesskette darzulegen. Durch diese Kriterien soll der in Ökobilanzen dargelegte Zielkonflikt zwischen der Minimierung der Treibhausgasemissionen und einer positiven ökologischen Gesamtbilanz möglichst aufgelöst werden (Zah et al., 2007)

Daten zur Biogasproduktion in Österreich und International

Die Errichtung und der Betrieb von Biogasanlagen erfolgten in Österreich durch stark wechselnde Rahmenbedingungen in den vergangenen Jahren nicht kontinuierlich. Im Ökostrombericht 2009 sind die aktuellen Statistiken zur Stromproduktion aus gasförmiger Biomasse in Österreich zusammengefasst. Mit 31.12.2008 waren 344 Anlagen mit einer Engpassleistung von 92 MW nach dem Ökostromgesetz anerkannt. Ein aufrechtes Vertragsverhältnis mit der Abwicklungsstelle für Ökostrom AG (OeMAG) bestand für 76,2 MW. Diese Anlagen wurden tatsächlich errichtet und sind in Betrieb. Die deutliche Differenz ist auf die Investitionsunsicherheit durch hohe Rohstoffpreise in den Jahren 2007 und 2008 zurückzuführen. Im Jahr 2008 erzeugten diese Anlagen 503 GWh Strom. Dies entspricht 11 % des kontrahierten Ökostroms und 1,0 % der inländischen Stromproduktion. Die durchschnittliche Anlagengröße beträgt 268 kWel (E-Control, 2009). Die Steigerung des Ausbaus seit 2001 ist in Abbildung 1 dargestellt.

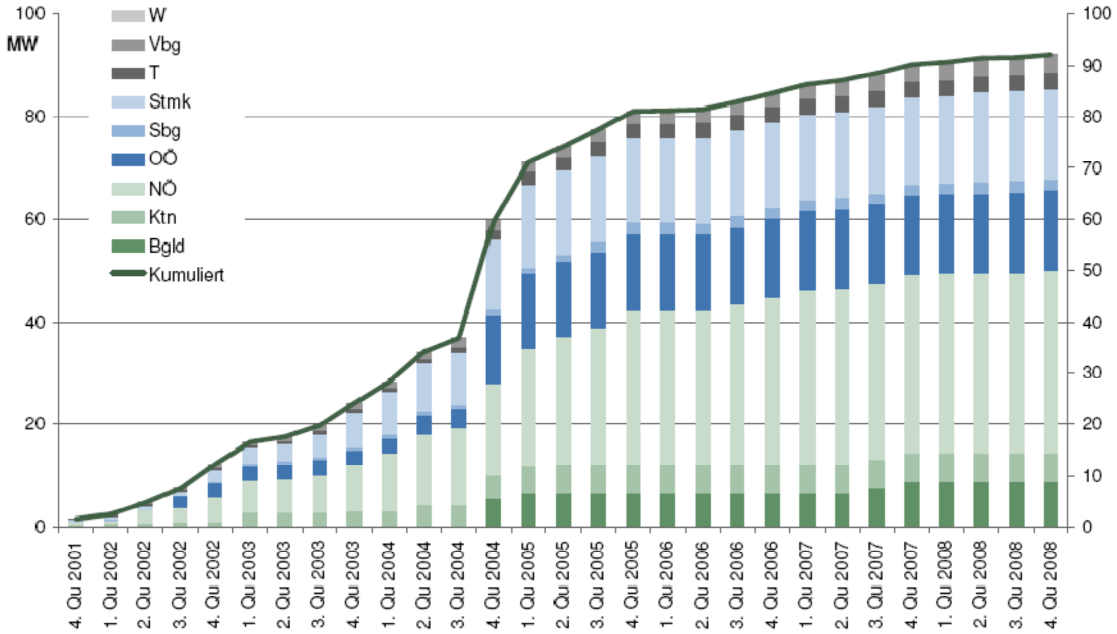

Abbildung 1: Entwicklung der Biogasanlagen in Österreich nach Ausbauleistung seit 2001 (E-Control, 2009)

In Abbildung 2 sind die eingesetzten Substrate nach ihrem energetischen Beitrag dargestellt. Besonders auffällig ist der Anteil von Maissilage mit 56,1 %. Abfälle und industrielle Nebenprodukte sind in dieser Darstellung nicht weiter aufgegliedert, da die Erhebung der Energie-Control GmbH auf die Mengen und Kosten eingesetzter landwirtschaftlicher Substrate fokussiert war.

Eine genauere Aufschlüsselung erlaubt die Anlagen- und Stoffdatenbank des Umweltbundesamtes. Diese weist 186 in Betrieb befindliche Anlagen aus, die zur anaeroben biotechnologischen Behandlung von Abfällen zugelassen sind. Diese Anlagen haben eine Kapazität von mehr als 393.000 Tonnen. Anlagen, die ausschließlich nachwachsende Rohstoffe (Silomais, Grassilage, Grünschnitt, Futterreste) verarbeiten, sind darin nicht enthalten (Umweltbundesamt, 2009).

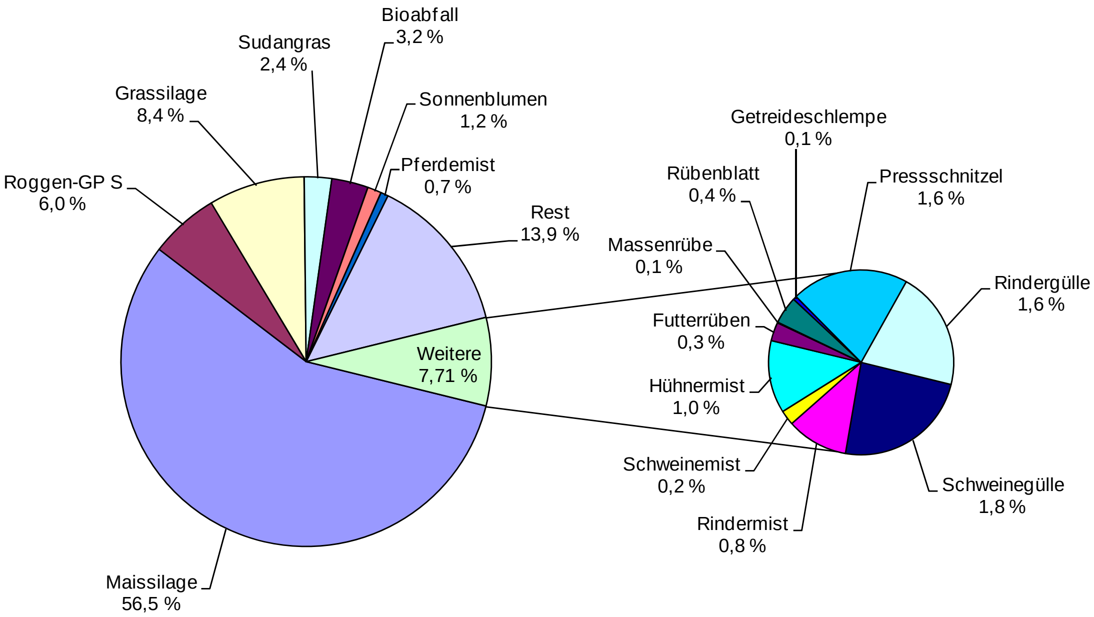

Abbildung 2: Eingesetzte Substrate in Biogas-Anlagen 2008 in Österreich nach Energieanteil (Sample 198 Anlagen) Quelle: Energie-Control GmbH

Die Verwendung des Biogases ist derzeit in Österreich stark von den Regelungen und den Einspeisetarifen für Ökostrom abhängig (Ökostromgesetz, 2009; Ökostrom-verordnung, 2010). Für die Biomethanproduktion wurden in der Energiestrategie Österreich und von Interessensvertretungen ambitionierte Ausbauziele genannt, die aber auch mit einer Neuorientierung des Vermarktungssystems verbunden sind und in einer Biomethanstrategie für den Zeitraum bis zumindest 2020 erarbeitet werden sollen (BMWFJ und BMLFUW, 2010; Kopetz, 2010).

In Deutschland wurde mit dem EEG 2000 und dessen Novellierungen 2004 und 2009 ein langanhaltender Ausbauboom bei landwirtschaftlichen Biogasanlagen erreicht. Zum Ende des Jahres 2009 waren in Deutschland etwa 4.500 Biogasanlagen mit einer installierten elektrischen Gesamtleistung von rund 1.650 MW in Betrieb. Das entspricht im Vergleich zum Jahr 2001, bezogen auf die installierte Leistung, dem rund 15-fachen (BMU, 2009).

In Europa wurden 2007 aus Biogas 19.938 GWh Strom produziert. Davon 9.520 GWh in Deutschland und 493 GWh in Österreich (AEBIOM, 2009).

Sowohl in Europa als auch in Österreich wird die Bioenergie eine wichtige Rolle bei der Erreichung der Ziele einnehmen (EurObserv´ER, 2009; IEA, 2009). Für die Biomethanproduktion berechnen die Interessensvertretungen ein Ausbaupotenzial auf 45,9 Mrd. m³ Biomethan. Dies entspricht der acht fachen Menge des Jahres 2007 (AEBIOM, 2009).

Energiedienstleistungen

Der große Vorteil von Biogas im Vergleich zu anderen Erneuerbaren Energien liegt vor allem auch darin, alle drei bedeutenden Endenergieformen Strom, Wärme und Treibstoff zur Verfügung stellen zu können. Die Produktion von Ökostrom dominiert sowohl in Österreich, in Deutschland die durch Biogas bereit gestellte Endenergie. Die Nutzung der Abwärme der Blockheizkraftwerke wurde bei der Gestaltung der jeweiligen Gesetze nur unzureichend gefordert. Die aktuellen Novellierungen drängen Anlagenbetreiber jedoch zu einem hohen Gesamtwirkungsgrad mit entsprechender technischer Ausstattung und Standortwahl.

Nach einer entsprechenden Aufbereitung zu Erdgasqualität wird ein etablierter Energieträger zur Verfügung gestellt, der durch die vorhandene Infrastruktur vielfältig eingesetzt werden kann. Verfahren zur Gasaufbereitung sind großtechnisch verfügbar, neue technologische Ansätze sind in Entwicklung. Biomethan als Treibstoff und als Beimengung zu komprimiertem Erdgas wird heute bereits in Schweden vor allem für kommunale Busflotten großtechnisch eingesetzt. In Deutschland und Österreich sind erste Demonstrationsprojekte umgesetzt.

Zusätzlich kann Biogas zeitlich und räumlich flexibel hergestellt werden. Eine nachfrageorientierte zur Verfügung Stellung der Energiedienstleistung ist somit möglich. Mit der Einbindung in das Erdgasnetz ist eine teilweise Neustrukturierung der Wertschöpfungskette verbunden, die von der Substratbereitstellung über den Betrieb der Anlage bis zur Einspeisung in das Erdgasnetz reicht. Neue Marktteilnehmer wie Gasversorger sind dadurch verstärkt an Bio-Methan interessiert. Je nach Substrat und Verarbeitungsweg müssen alle Optimierungsschritte genutzt werden.

3.3. Herausforderungen und neue Entwicklungen

Den Vorteilen der Verwertung von organischen Abfällen, industriellen Nebenprodukten, landwirtschaftlichen Reststoffen und Energiepflanzen in anaeroben Prozessen zur Erreichung von vielfältigen Umwelt- und Energiezielen steht durch die Komplexität des Systems eine Vielzahl von Herausforderungen gegenüber, die in neuen Entwicklungen gelöst werden müssen.

Die Verfahrenskette von den Substraten bis zur Gasverwertung ist durch ein Ineinandergreifen von verschiedenen Technologien und naturwissenschaftlichen Disziplinen gekennzeichnet. Mechanische, chemisch-physikalische und biotechnologische Fragestellungen müssen für sich und im Zusammenwirken optimiert werden (Pind et al., 2003).

Ahring (2003) sieht als wichtige zukünftige Werkzeuge, biotechnologische Forschung und Molekulartechnologien, um dynamische Studien über den Prozessablauf unter industriellen Bedingungen erstellen zu können. Die Forschung wird sich dabei auch auf konkrete Anwendungsfälle und Großanlagen konzentrieren. Daraus leitet sie die Forschungsschwerpunkte der Zukunft ab, bei denen Fragestellungen der Grundlagenforschung und anwendungsbezogene Forschung und Entwicklung zusammen wirken müssen.

Sie gliedert die Entwicklungsschwerpunkte für die Anaerobtechnologie in vier Bereiche:

1. Verbesserung der Abbaubarkeit der Substrate
2. Optimierung der Verfahrenstechnik der Anlagen
3. Optimierung der Prozesskontrolle
4. Verbesserung des mikrobiologischen Prozesses und Steigerung der Effizienz

Holm-Nielsen et al. (2009) ergänzt diese Punkte um die Notwendigkeit einer verbesserten Verwertung der Gärreste.

Weiland zieht aus dem zweiten bundesweiten Messprogramm bei 61 landwirtschaftlichen Biogasanlagen in Deutschland, die vor allem für die Erzeugung Erneuerbarer Energie ausgelegt wurden, folgende Schlüsse für zukünftige Entwicklungsschritte der Energiepflanzenvergärung (FNR, 2010; Weiland, 2010):

1. Anpassung der Anlagentechnik und der Betriebsweise an die stofflichen Anforderungen der Substrate
2. Sorgfältige Anlagenplanung und Standortwahl, die eine effiziente Biomassebereitstellung und ein sorgfältig ausgearbeitetes Nutzungskonzept für das produzierte Gas beinhaltet.
3. Störungsarmer Betrieb und Verminderung von Energieverlusten entlang der gesamten Wertschöpfungskette
4. Effiziente Ausnutzung der eingesetzten Substrate

Aus den vielen Anwendungsfällen der Anaerobtechnologie und den daraus ableitbaren Optimierungsansätzen wurden im Rahmen dieser Arbeit zwei Prozessketten genauer untersucht. Sowohl bei der Verwertung von Reststoffen als auch bei der Nutzung von Energiepflanzen sind die jeweilige ökonomische Optimierung und die Einbettung in das Zielsystem Umwelt, Energie, Klimaschutz von Bedeutung.

2. Problemstellungen und Forschungsziele

In den verfassten Publikationen werden Lösungsansätze zu aktuellen Problemen in zwei Anwendungsgebieten der Biogasproduktion erarbeitet. Die bestehenden Herausforderungen und vorgeschlagenen Optimierungen werden in das dargestellte Zielsystem Umwelt, Energie, Klimaschutz eingebettet:

- Aus dem Bereich der Reststoff- und Nebenproduktverwertung werden aktuelle Fragstellungen in der Co-Vergärung und in der Mono-Vergärung von Schlachtabfällen und anderer problematischer Substrate der Lebensmittel-industrie und der Biotreibstoffproduktion bearbeitet. Mit diesen Substraten sind durch den konzentrierten Anfall in Produktionsanlagen besondere Herausforderungen verbunden.

  Es wird ein neuer Verfahrensvorschlag präsentiert, der eine Stabilisierung des Prozesses und eine Minimierung von Hemmungen durch freien Ammoniak und die Akkumulation freier flüchtiger Fettsäuren ermöglicht.

  Ein weiterer Fokus der durchgeführten Untersuchungen liegt in der weiteren Behandlung des Gärrests. Durch die präsentierten Lösungsansätze soll eine bessere Integration der Reststoffverwertung und der damit verbundenen Produktion Erneuerbarer Energie in den industriellen Produktionsprozess erreicht werden.

  [Publikationen 1, 3, 4, 6, 7, 8]

- Den zweiten Schwerpunkt bildet die Vergärung von Energiepflanzen als feste Substrate ohne die Beigabe von Gülle. Diese neue Anwendungsmöglichkeit der Biogastechnologie zur Produktion Erneuerbarer Energie ist mit Fragestellungen zur Ökobilanz und zu den Produktionskosten der Energiepflanzen konfrontiert.

  Durch die Bilanzierung einer Großanlage mit ausschließlicher Energiepflanzen-vergärung über 20 Monate sollen die Auswirkungen auf die Prozessführung bei unterschiedlichen Substratmischungen dargestellt werden. Verfahrens-technische Lösungsvorschläge zur Optimierung des Ausgärgrades wie die Mengeneinstellung der Rezirkulationsflüssigkeit oder die Reduzierung der festen abgepressten Gärrestmenge werden aufgezeigt. An einer anderen in Betrieb befindlichen Anlage werden die Auswirkungen einer Leistungssteigerung durch mehr Substratzugabe auf den Ausgärgrad des Gärrests untersucht.

  [Publikationen 2, 5]

3. Ergebnisse und Diskussion

3.1. Lösungsansätze zur Reststoff- und Nebenproduktverwertung problematischer Substrate

Die Verwertung von Abfällen und Nebenprodukten ist eine der wichtigsten Aufgabenstellungen des Umweltschutzes in den letzten Jahren. Durch die Hierarchie in der Abfallwirtschaft und dem betrieblichen Umweltschutz, welche die Vermeidung vor der Verwertung und erst an dritter Stelle die Entsorgung sieht, haben biologische Verwertungstechnologien gegenüber der Deponierung und der Verbrennung an Bedeutung gewonnen. Die Anaerobtechnologie ist durch die Nutzungsmöglichkeit des Gases im Grünbuch über die Bewirtschaftung von Bioabfall in der Europäischen Union als Verwertungstechnologie anerkannt (Europäische Kommission, 2008). Ein zusätzlicher Vorteil ist die Rückgewinnung der Nährstoffe und die Verwendung als landwirtschaftlicher Dünger unter Einhaltung von Qualitätskriterien (Hartmann und Ahring, 2006). Entwicklungs- und Optimierungsschritte müssen sowohl entlang der Prozesskette der Biogasanlage als auch bei der Einbindung in die Abfallwirtschaft oder den jeweiligen Produktionsbetrieb und auf der Seite der Gasverwertung in das neue Energiesystem erfolgen. Für das Anwendungsfeld der Abfall- und Nebenproduktverwertung sind die wichtigsten identifizierten Optimierungsthemen in Abbildung 3 aufgelistet.

Weltweit wird die Vergärung von heterogenen organischen Abfällen aus Siedlungen und Gewerben großtechnisch eingesetzt. Die Zusammensetzung der genutzten Abfälle und notwendige Aufbereitungstechnologien sind dabei von der Siedlungsstruktur und dem lokalen Abfallwirtschaftssystem abhängig (Loll, 2002). Die Zusammensetzung der organischen Fraktion, aber auch die enthaltenen Stör- und Hemmstoffe beeinflussen das Design und die Prozessführung der Vergärungsanlage, wie zum Beispiel die notwendigen Aufbereitungs- und Trennungsschritte (Edelmann und Engeli, 2005).

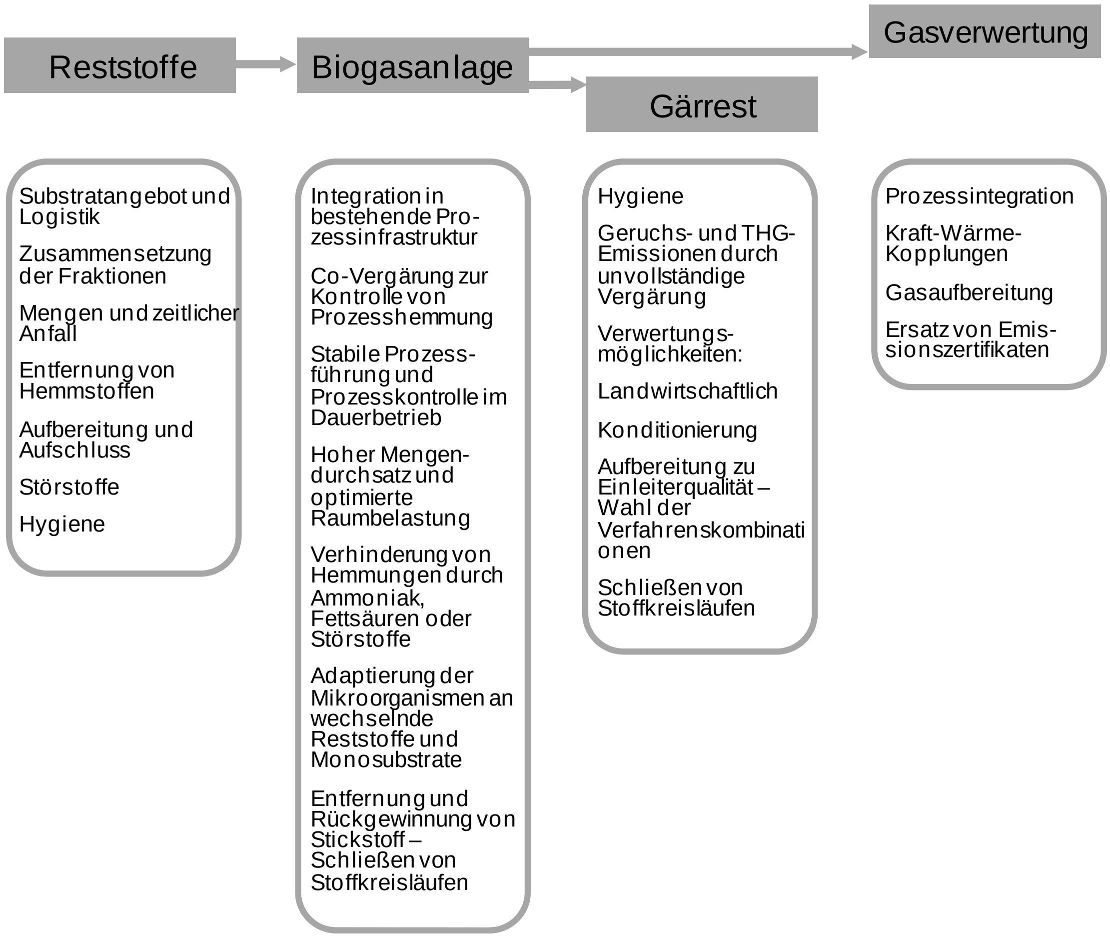

Abbildung 3: Themen für zukünftige Optimierungen bei Biogasanlagen aus der Abfall- und Nebenproduktverwertung

Die Reststoffe und Nebenprodukte aus der Lebensmittelindustrie und andere nasse organische Reststoffe von Märkten oder Großküchen wurden lange in einem lokalen Stoffkreislauf mit der Landwirtschaft verwertet. Die Konzentration der Unternehmen der Lebensmittelindustrie führt heute zu großen Mengen produktspezifischer Nebenprodukte. Durch den Strukturwandel in der Landwirtschaft und die notwendigen Einschränkungen durch gesetzliche Regelungen konnten die Nebenprodukt- und Nährstoffkreisläufe nicht weiter aufrecht gehalten werden (Russ und Schnappinger, 2007). Die anaerobe Verwertung und die Integration der Energiegewinnung in die Produktion bieten neue Chancen, die Stoffkreisläufe in einem größeren Rahmen sicher zu stellen und die Energieproduktivität des Produktionsprozesses zu erhöhen. Der Einsatz von Primärenergie je produzierter Einheit kann gesenkt werden.

Bei der Nutzung in Biogasanlagen stellt die Verwendung von organisch hoch-konzentrierten Reststoffen aus der Lebensmittelindustrie, der Lederfabrikation und der Biotreibstoffproduktion eine besondere Herausforderung dar. Der hohe Anteil an Proteinen und Fetten bei einigen der verfügbaren Reststoffe- und Nebenprodukte ist bei einer Mono-Vergärung dieser problematischen Substrate Ursache für die Hemmung durch freien Ammoniak und die Limitierung der mikrobiologischen Abbauleistung durch langkettige Fettsäuren:

Beim Abbau von Proteinen über Aminosäuren wird der enthaltene Stickstoff als Ammoniumion freigesetzt. Abhängig von pH Wert und Temperatur bildet Ammonium ein Dissoziationsgleichgewicht mit Ammoniak. Undissozierter Ammoniak wird durch die Zellmembran von den Mikroorganismen aufgenommen, hemmt deren Stoffkreislauf und damit den Abbau der organischen Substanzen. Die Auswirkungen auf den Abbauprozess wurden in der Literatur unter anderem von McCarty and McKinney (1961), van Velsen (1979), Braun et al. (1981), Hashimoto (1986), Wiegant und Zeeman (1986), Angelidaki und Ahring (1993), Hansen et al. (1998), Salminen und Rintala (2002) Chen et al. (2008) sowie von Schnürer und Nordberg (2009) dokumentiert. Eine verlangsamte und reduzierte Abbauleistung, die Akkumulation von freien flüchtigen Fettsäuren und in weiterer Folge ein Absinken des pH Wertes sind die wichtigsten Auswirkungen.

Der Abbau von Fetten über langkettige Fettsäuren limitiert das Zusammenwirken durch eine Anlagerung der Moleküle an die Konsortien der Mikroorganismen. Als Auswirkung wurden eine reduzierte Abbauleistung und eine Anreicherung freier flüchtiger Fettsäuren beobachtet (Lokshina et al., 2002; Peirera et al., 2004; Cirne et al., 2007).

3.1.1. Co-Vergärung als Lösungsansatz zur Kontrolle von Prozesshemmungen

Die negativen Auswirkungen zu hoher Stickstoffkonzentrationen oder Fettanteile auf den mikrobiologischen Prozess können durch eine geeignete Auswahl der eingesetzten Substrate minimiert werden. Durch die Zusammensetzung des Substratmixes kann die Konzentration der potentiell hemmenden aktiv gesteuert werden und die Versorgung mit Nährstoffen und Spurenelementen optimiert werden. Bei der kontinuierlichen Führung des Prozesses ist dazu eine genaue analytische Kontrolle wichtiger Parameter erforderlich.

Die Co-Vergärung mehrerer Abfallströme ermöglicht eine hohe Auslastung beim Betrieb industrieller Biogasanlagen. Dadurch können die Betriebskosten reduziert werden. Die Erlöse können durch die Annahme unterschiedlicher Stoffströme maximiert werden. Diesen ökonomischen Vorteilen stehen die erhöhten Errichtungskosten durch mehr maschinelle Infrastruktur gegenüber (Mata-Alvarez et al., 2000).

Eine Vielzahl von Substratkombinationen und Prozesstechnologien sind möglich und wurden von Hartmann und Ahring (2006) zusammenfassend dargestellt. Sie berichten, dass das Verhältnis von abbaubarem Kohlenstoff zum Stickstoffgehalt (C:N Verhältnis) durch die Wahl der Substrate in einem Bereich von 25 bis 30 eingestellt werden soll, um einerseits eine ausreichende Nährstoffversorgung sicher zu stellen und andererseits das Risiko einer Hemmung zu minimieren. Sie kommen zu dem Schluss, dass ein hoher Biogasertrag, vor allem auch durch eine gute Feststoffabtrennung und Aufbereitung der Substrate, erreicht werden kann.

Salminen und Rintala (2002) fassen die Verwertungsoptionen an Hand der anaeroben Vergärung von Nebenprodukten der Hühnerschlachtung zusammen. Am geeignetsten zur anaeroben Verwertung sehen sie das Blut und das Fett aus Abscheidern. Edström et al. (2003) können in Pilotversuchen durch die Zugabe von Schlachtabfällen zu anderen organischen Abfällen die Biogasausbeute deutlich steigern. In kontinuierlichen Versuchen beobachten sie Adaptierung der Mikroorganismen an einen höheren Stickstoffgehalt selbst bei hoher organischer Raumbelastung.

In eigenen halbtechnischen kontinuierlichen Versuchen wird die Entscheidungsgrundlage zur Erweiterung einer industriellen Co-Vergärungsanlage aufbereitet. Durch die Beimengungen von Reststoffen aus der Lebensmittelindustrie kann die Biogasausbeute deutlich gesteigert werden. Für die Betreiber der Anlage ist entscheidend, eine größere Menge an Reststoffen behandeln zu können, um damit die Anlage besser auszulasten. Es kann gezeigt werden, dass durch eine Co-Vergärung von nassen organischen Siedlungsabfällen mit Reststoffen aus der Lebensmittelindustrie (Fettfraktionen und Hühnerblut) bei hohen Raumbelastungen (10 kg_{CSB} m^3 d^{-1}) eine hohe Abbaurate erzielbar ist Eine weitere Erhöhung der Raumbelastung auf 12 kg_{CSB} m^3 d^{-1} und des Stickstoffgehalts durch Erhöhung des Anteils von Fett und Blut führen jedoch zu einer Anreicherung von freien flüchtigen Fettsäuren und zu einer Verschlechterung der Abbauleistung. Zur Stabilisierung des Vergärungsprozesses und zur Steigerung der Abbauleistung wird die Einsatzmöglichkeit einer zweiten Vergärungsstufe durch Batch-Gärversuche mit dem vorliegenden Gärrest evaluiert (Resch et al., 2006) [Publikation 1].

Die Erweiterung um eine Vergärungsstufe sichert die Einhaltung von Umweltauflagen, wie den Abbau der organischen Substanzen und die Minimierung von Geruchsemissionen. Durch die praxisnahen Versuche kann gezeigt werden, dass die Co-Vergärung der vorliegenden Fraktionen auch im industriellen Maßstab eine technisch und wirtschaftlich sinnvolle Kombination ist, wenn die hemmenden Einflüsse aus den tierischen Nebenprodukten durch die Wahl der Substratzusammensetzung wie das Einstellen des C:N Verhältnisses aktiv kontrolliert werden können.

3.1.2. Mono-Vergärung von tierischen Nebenprodukten

Eine Co-Vergärung mit anderen Reststoffströmen ist nicht immer möglich und oft auch nicht gewünscht. Dies ist der Fall, wenn die Reststoffverwertung integriert in den Standort des Produktionsbetriebs erfolgen soll, um die Energie des Biogases zur Deckung des Eigenenergieverbrauchs zu nutzen. Bei einem Schlachthof ist diese Integration besonders vorteilhaft: Im Schlachtbetrieb ist neben elektrischer Energie vor allem Wärme in mehreren Temperaturniveaus notwendig. Dies reicht von Warmwasser zur Reinigung bis zu Heißwasser und Prozessdampf.

Typischerweise wird das Biogas in einem Blockheizkraftwerk (BHKW) zu Strom und Wärme umgewandelt, die als Nutzenergien im Schlachthof bereit gestellt werden können. Es ist aber auch die Aufbereitung des Biogases und damit die direkte Substitution von Erdgas oder die Bereitstellung von Prozessdampf möglich. Die Wärmenachfrage im Schlachthof ist an die Betriebszeiten gebunden, wohingegen die Wärme aus dem BHKW kontinuierlich bereit gestellt wird. Durch eine standortspezifische Integration ist eine ganzjährige Verwendung der bereitgestellten Energie erreichbar. Kirchmayr et al. (2009) demonstrierten eine Abdeckung von 50 % des Gesamtenergiebedarfs vor allem durch eine bessere Prozesseinbindung der Abwärmenutzung durch die Installation eines Warmwasserspeichers bei einem Schlachthof in Oberösterreich. Nahezu die gesamte Abwärme des BHKWs kann genutzt werden, um 80 % des Wärmebedarfs des Schlachthofes abzudecken.

Diese Erweiterung hat Beispielwirkung für andere Anwendungsfälle, da mit der Substitution des fossilen Primärenergieträgers Erdgas einerseits und mit der Abwärmenutzung aus der Stromproduktion des BHKWs andererseits ein Beitrag zur Erreichung der Energie- und Klimaziele im Bereich der Industrie erreicht wird.

Basis für diese sinnvolle Prozessintegration ist ein stabiler und funktionierender anaerober Abbauprozess in der Biogasanlage. Kirchmayr et al. (submitted) [Publikation 7] beschreiben die langjährige Optimierung des Anaerobprozesses. In dem betrachteten Schweineschlachthof fallen je Schlachtkörper etwa 5 Liter Blut, 6 Liter Darm und Darminhalt an, die gemeinsam mit 10 Liter Fett eines Abscheiders in der Biogasanlage verarbeitet werden. Eine Zusammenfassung der chemischen Zusammensetzung und der erreichbaren Methanausbeuten einzelner Fraktionen werden im Buchbeitrag von Kirchmayr et al. (2007) [Publikation 4] präsentiert.

Der limitierende Faktor bei der Monovergärung dieser Substrate ist der hohe Stickstoffgehalt der proteinreichen tierischen Nebenprodukte. Der durch den hohen Ammonium- bzw. Ammoniakgehalt gehemmte mikrobiologische Prozess führt zu einem unvollständigen Abbau der organischen Substanzen und zur Akkumulation von organischen Säuren von mehr als 10 g l^{-1}. Kann der Fermenterinhalt bis zum Endlager nicht stabilisiert werden, werden konzentrierte Geruchsemissionen an der Anlage messbar. In weiterer Folge entstehen bei der unbehandelten Ausbringung des Gärrests als Stickstoffdünger auf landwirtschaftliche Flächen unangenehme Geruchs-emissionen.

Ein aus der Perspektive der Nutzung Erneuerbarer Energie und aus der Perspektive der Abfallwirtschaft sinnvolles Verfahren - nämlich die Verwertung von Reststoffen - gerät durch diese Geruchsemissionen und die Gefahr der Grundwasserbelastung durch Überdüngung mit Gärrest in Konflikt mit zentralen Umweltschutzzielen. Eine Möglichkeit Geruchsemissionen beim Ausbringen von Gärresten zu vermeiden, ist die Festlegung von Qualitätsstandards. Wimmer (2005) bezieht sich in seinen Schlussfolgerungen zur Reduzierung der Geruchsemissionen bei der Ausbringung auf den Erlass des niedersächsischen Umweltministeriums zum Immissionsschutz bei Biogasanlagen (MU 33 – 40501/208.13/1, 2004), der einen Gesamtgehalt organischer Säuren von kleiner 2 g l^{-1} bei der Ausbringung vorschreibt.

Bei der betrachteten Biogasanlage sind daher eine genaue Prozessführung, laufende Prozesskontrolle an der Anlage und ergänzende Laboruntersuchungen notwendig, um einen stabilen und zufriedenstellenden Prozess zu gewährleisten. Die Auswertungen von Kirchmayr et al. (submitted) [Publikation 7] zeigen, dass derzeit nur ein Teil der zur Verfügung stehenden tierischen Nebenprodukte in der Anlage verwertet werden kann. So müssen zum Beispiel über den Jahresschnitt gesehen 50 % des Blutes auf Grund des hohen Proteingehaltes in der Tierkörperverwertung entsorgt werden.

3.1.3. Stickstoffentfernung und Rückgewinnung aus dem Biogasprozess

In den durchgeführten Projekten war neben der Prozessstabilisierung vor allem auch die Kapazitätserweiterung für zusätzliche tierische Nebenprodukte, vor allem Blut, Ziel der Arbeiten. Eine Möglichkeit den Durchsatz tierischer Nebenprodukte in der Biogasanlage des Schlachthofes zu erhöhen, ohne die Umweltschutzziele zu verfehlen, ist die Entfernung des Stickstoffs aus dem Prozess. Ein niedrigerer Stickstoffgehalt im Fermenter reduziert das Risiko einer Prozesshemmung durch freien Ammoniak. Eine Rückgewinnung des Stickstoffs in nutzbarer Form verbessert zusätzlich das Dünge- und Nährstoffmanagement.

Durch die Intensivierung der Landwirtschaft wird der natürliche Stickstoffkreislauf maßgeblich durch den Menschen beeinflusst. Die Düngerproduktion durch das Haber-Bosch Verfahren hat dabei den größten Anteil. 2008 wurden nach Angaben des US Geological Service (USGS) 136 Millionen Tonnen Stickstoffdünger unter hohem Druck und hoher Temperatur synthetisiert (USGS, 2009). Dabei werden knapp 2 % des weltweiten fossilen Energieeinsatzes verbraucht (IEO, 2010). Der anthropogene Stickstoffkreislauf ist in Abbildung 4 dargestellt.

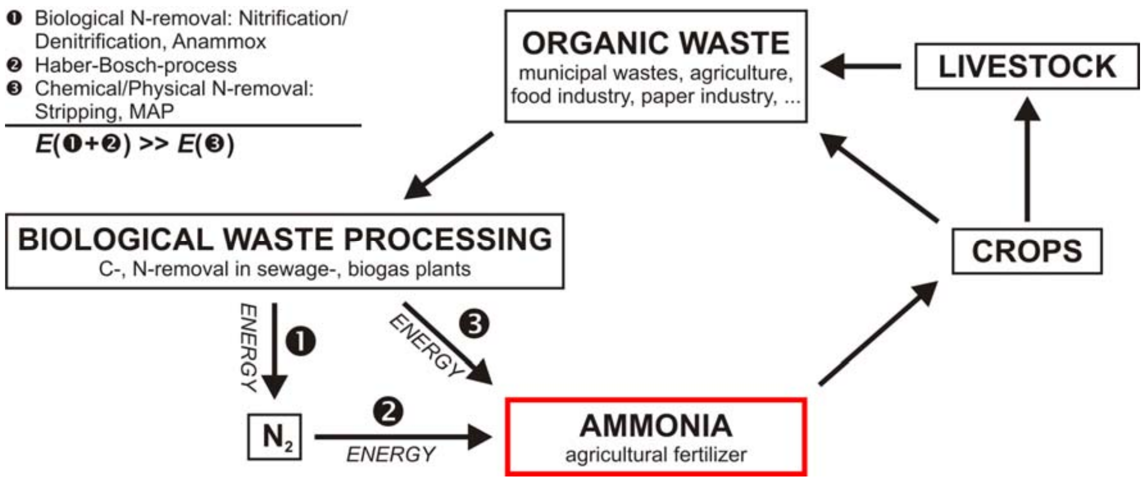

Abbildung 4: Anthropogener Stickstoffkreislauf nach Wörl (2007): Der in der Organik von Reststoffen gebundene Stickstoff wird neben dem diffusen Eintrag in Böden und Gewässer größtenteils in biologischen Abwasserreinigungsanlagen durch Nitrifikation und Denitrifikation entfernt und als N_2 an die Atmosphäre abgegeben. (1) Durch das energieintensive Haber-Bosch Verfahren wird der Luftstickstoff N_2 wieder zu Ammoniak und in weitere Folge zu Mineraldüngerprodukten synthetisiert (2). Die Entfernung und Rückgewinnung durch physikalische und chemische Methoden aus Verwertungsprozessen kann den Energieeinsatz deutlich minimieren (3). N = Stickstoff, E = Energie

Die Anaerobtechnologie kann durch den sachgerechten Einsatz des Gärrests in der Landwirtschaft einen wichtigen Beitrag beim Schließen von Stickstoffkreisläufen leisten. Bei der Vergärung von landwirtschaftlichen Reststoffen wie Gülle ist dies etabliert. Die Vergärung von organischen Siedlungsabfällen und die angeschlossene Verwendung des Gärrests (z.B. Kompostierung) werden ebenso großtechnisch eingesetzt. Die Verwertung konzentrierter Reststoffe und Nebenprodukte aus der Lebensmittelindustrie stellt durch den hohen Stickstoffgehalt noch eine Herausforderung dar.

Bei hohen Stickstoffgehalten im Fermenterinhalt bzw. im Gärrest kann es zur Steigerung der Abbauleistung im Vergärungsprozess und zur Steigerung der Gärrest-qualität sinnvoll sein, den Stickstoff gezielt zu entfernen.

Zur Entfernung und Rückgewinnung von Stickstoff aus Vergärungsanlagen stehen Verfahren zur Strippung von Ammoniak zur Verfügung. Beim Strippen wird Ammoniak aus dem flüssigen Fermenterinhalt durch das Durchleiten von Gasen (wasserdampfgesättigte Luft oder Wasserdampf) aus der Flüssigkeit entfernt und in das Gas übergeführt. Die treibende Kraft hinter diesem Prozess ist, dass der Dampfdruck der aus der Flüssigkeit zu entfernenden Stoffe in der Flüssigkeit größer als vorbeiströmenden Gas ist und daher ein Übertritt von der Flüssigkeit in das Gas erfolgt. Das chemische Gleichgewicht von Ammonium und Ammoniak wird von der Temperatur und dem pH Wert beeinflusst. Der Stoffübergang beim Strippen beruht auf dem Henry´schen Gesetz, das von der Temperatur, den Stoffeigenschaften und dem umgebenden Partialdruck bestimmt wird. (Sattler, 2001)

In der Literatur sind verschiedene Verfahren beschrieben (Bonmati und Flotats, 2003; Siegrist et al., 2005; Angelidaki et al., 2006). Bei diesen wird der Stickstoff im Anschluss an die Fermentation aus physikalisch aufbereitetem Gärrest gestrippt und das stickstoffreduzierte Prozesswasser rezirkuliert. Der Vorteil dieser Anordnung ist, dass aus der chemischen Industrie bekannte Strippkolonnen für die umwelttechnische Anwendung adaptiert werden können. Die Nachteile liegen in der größeren Flüssigkeitsmenge, die wegen der Rezirkulation laufend anaerob behandelt und für die Strippung aufbereitet werden müssen. Bei einer Integration in eine bestehende Anlage müssten daher zusätzliche Fermentervolumina errichtet werden oder bei gleichbleibender Abfallmenge die hydraulische Aufenthaltszeit in den Fermentern reduziert werden.

Aus diesen Überlegungen sollte ein Verfahren entwickelt werden, das eine Entfernung des Stickstoffs direkt aus dem anaeroben Vergärungsprozess ermöglicht und bei der Vergärungsanlage am Schlachthof eingesetzt werden kann. Abbildung 5 illustriert diesen Verfahrensablauf und die Unterschiede bei den Massenflüssen im Vergleich zur nachträglichen Entfernung. Zeng (2006) und Nakashimada (2008) haben vergleichbare Verfahrensansätze präsentiert.

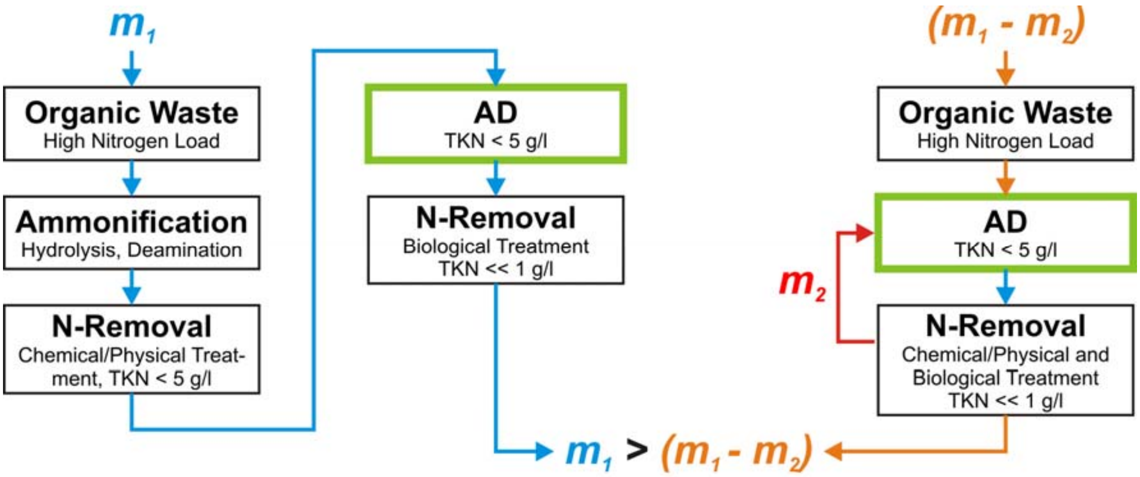

Abbildung 5: Die Entfernung von Stickstoff durch nachgeschaltete Strippung (rechts) und die Entfernung direkt aus dem Fermentationsprozess (links) mit eventuell nachgeschalteter biologischer Aufbereitung des Gärrests (Mitte). Der Vorteil des integrierten Prozesses ist der größere Massenfluss m1 im Vergleich zur Rezirkulation des Gärrests (m1 – m2) (Wörl, 2007)

Um das vorgeschlagene Verfahren wie in Abbildung 5 zu verwirklichen, müssen zumindest drei Prozessschritte entwickelt oder optimiert werden:

1. Vorab müssen die Proteine in den tierischen Nebenprodukten zu Aminosäuren hydrolisiert werden. In einer Desaminierung muss mit Hilfe der Sticklandreaktion das Ammonium aus hydrolisierten Aminosäuren abgespalten werden (Ramsay und Pullammanapallil, 2001; Griehl et al., 2002). Versuche mit unterschiedlichen Enzymen und der Inokulation von Blut mit C. *sticklandii* wurden von Unterlerchner (2006) durchgeführt. Die grundsätzliche Eignung konnte gezeigt werden. Die nachgeschalteten Prozessschritte sollen aber vorrangig weiterentwickelt werden, da die Freisetzung von Ammonium auch „unkontrolliert“ in einem ersten vollständigen anaeroben Fermentationszyklus realisiert werden kann.

2. Ein robuster Prozess muss entwickelt werden, der die Entfernung des Ammoniums aus dem Fermenterinhalt ermöglicht. Um die weiteren mikrobiologischen Reaktionen möglichst wenig zu beeinflussen, soll dabei auf die Zugabe von Laugen zur Steigerung des pH verzichtet werden. Da der Ausgangs pH durch den hohen Stickstoffgehalt im Fermenter > 8,1 ist, wurde für die Labortests ein Dampfstrippverfahren gewählt. Eine Pilotanlage, die einen kontinuierlichen Stripp-Prozess erlaubt, wurde im Technikumsmaßstab (120 $l\ h^{-1}$) konstruiert. Die Versuchsergebnisse wurden von Wörl (2007) und später von Bock (2008) zusammengefasst. Es konnte gezeigt werden, dass der Stickstoffgehalt im gestrippten Fermenterinhalt um 67 % auf 2,5 $g\ l^{-1}$ gesenkt werden konnte.

3. Die Auswirkungen der Entfernung des Ammonium-Stickstoffs auf eine Verbesserung des Fermentationsprozess wurden in einer Reihe von kontinuierlichen Abbauversuchen im Labormaßstab getestet. Dabei wurden unterschiedliche zukünftige Betriebszustände der betrachteten großtechnischen Biogasanlage simuliert und mit den entsprechenden Referenzsituationen der Vergärungsanlage am Schlachthof verglichen (Resch et al., 2007a; Resch et al., submitted [Publikation 3]). Durch eine Reduktion des Gesamtstickstoffs auf 4 $g\ kg^{-1}$ wird der Abbau der organischen Substanz um 40 % deutlich gesteigert. Vor allem der nahezu vollständige Abbau der organischen Säuren unter 0,67 $g\ l^{-1}$ stabilisiert den Prozess. Bei der Simulation zukünftiger Betriebszustände konnte gezeigt werden, dass bei zufriedenstellender Abbauleistung nach der Implementierung der Stickstoffentfernung entweder die organische Raumbelastung durch den Einsatz zusätzlicher tierischer Nebenprodukte wie Blut um 60 % gesteigert werden könnte oder die hydraulische Aufenthaltszeit in der Anlage durch mehr Input um 25 % gesenkt werden kann. Als zufriedenstellende Abbauleistung wurde bei einem Abbau der verfügbaren Organik von zumindest 80 % und einer maximalen Konzentration von organischen Säuren im Gärrest von 1,5 $g\ l^{-1}$ definiert.

Die Versuchsergebnisse bereiten die Entscheidung für eine Weiterentwicklung des skizzierten Verfahrens auf. Ein nächster Schritt sollte die verfahrenstechnische Weiterentwicklung der Entfernung und der Rückgewinnung des Stickstoffs direkt aus dem Fermenterinhalt mit dem Ziel der Implementierung an der Großanlage sein. Bei der Integration an der Großanlage ermöglicht die Abtrennung des Stickstoffs und die Rückgewinnung als konzentrierter Stickstoffdünger in weiterer Folge eine kosteneffiziente Erweiterung des Einsatzradius bei der Ausbringung auf landwirtschaftliche Flächen. Waltenberger et al. (2007) berechnen, dass durch die Entfernung und Rückgewinnung des Ammoniums 55 % der Stickstoffmenge als konzentrierter Dünger auf überregionalen Flächen ausgebracht werden können.

Die Versuche zum Verfahren der Stickstoffentfernung tragen dazu bei, die Ziele der Abfallwirtschaft und Umweltschutzes besser umzusetzen. Durch die definierten Gärrestfraktionen kann die Stickstoffmenge genauer dosiert und an die lokalen Gegebenheiten angepasst ausgebracht werden. Dadurch wird die Aufnahme des Düngers durch die Pflanzen optimiert und die Auswaschung in das Grundwasser minimiert. Der Fachbeirat für Bodenfruchtbarkeit und Bodenschutz legt für Österreich den sachgerechten Einsatz von Biogasgülle und Garrückständen im Acker- und Grünland fest (BMLFUW, 2007b). Die anaerobe Verwertung von Reststoffen in Kombination mit der Rückgewinnung von definiertem Stickstoffdünger schließt den Nährstoffkreislauf und reduziert den Einsatz von energieintensiv produziertem Mineraldünger.

Zusätzlich kann durch die größere Menge an behandelbaren Nebenprodukten nach der Implementierung einer Stickstoffentfernung auch die Erzeugung Erneuerbarer Energie gesteigert werden. Bei der Erreichung der Energie- und Klimaziele im Rahmen der Verpflichtungen gegenüber der EU sind integrierte Industrielösungen von besonderer Bedeutung. Der effiziente Einsatz von Primärenergie in Produktionsprozessen und das Nutzen von Abwärmepotenzialen leisten einen wichtigen Beitrag, müssen aber auf die individuelle Situation ausgelegt werden.

3.1.4. Weitergehende Gärrestaufbereitung

Bei einer Integration der Biogasanlage in einen Betrieb mit großen Mengen organischer Reststoffe oder Nebenprodukte kann die regionale Verwertung des anfallenden Gärrests auf landwirtschaftlichen Flächen nicht mehr realisiert werden. Dies ist zum Beispiel der Fall, wenn eine Biogasanlage in den Prozess einer zentralen Bioethanol- oder Biodieselanlage integriert werden soll.

Am Beispiel der Produktion von Bioethanol wird deutlich, dass der hohe Einsatz von Prozessenergie zur Trocknung der Schlempe die Energiebilanz erheblich verschlechtert. Dies hat auch Einfluss auf die Ökobilanz und den netto zu erreichenden Beitrag zu den Klimaschutzzielen (von Blottnitz, 2007; Zah et al., 2007).

In einer Potenzialstudie wurden am Beispiel der Bioethanolanlage Pischelsdorf im Vergleich zum etablierten Weg der Trocknung und Futtermittelproduktion aus der Schlempe alternative Varianten untersucht. Das Konzept sieht vor, Teilströme der anfallenden Schlempe in einer Biogasanlage zu verwerten und das produzierte Biomethan in das Energiesystem der Bioethanolanlage zu integrieren. Drosg et al. (2008) [Publikation 6].

Neben prozesstechnischen Herausforderungen, wie dem hohen Stickstoffgehalt der Schlempe und der mögliche Mangel an Spurenelementen, muss vor allem für den Gärrest eine großtechnisch anwendbare Lösung gefunden werden. Die lokale landwirtschaftliche Verwertung steht nicht zur Verfügung, da bei einer Produktionsmenge von 200.000 t a^{-1} Bioethanol 1.400.000 t a^{-1} Schlempe entstehen. Für eine sachgerechte landwirtschaftliche Verwertung wären 36.000 ha, große Speichervolumina und eine immense Logistik notwendig. Dies bedeutet, dass eine Aufbereitung des Gärrests auf Einleiterqualität realisiert werden müsste. Die Berechnungen ergeben, dass 50 % der Kosten der Biogasproduktion für die Aufbereitung des Gärrests aufgewendet werden müssten. Als günstigstes Szenario wird in der Studie der Einsatz eines Membranverfahrens nach Vorkonditionierung vorgeschlagen.

Alternativ zum Membranverfahren können aerobe biologische Reinigungsverfahren zur Nitrifikation und Denitrifikation implementiert werden (Del Pozo et al., 2003; Fu et al., 2008, Mayer et al., 2009). Fuchs et al. [Publikation 8] zeigen dazu in früheren Versuchen, dass die Effizienz des Reinigungsprozesses durch den Einsatz von Filtergewebe zur Schlammrückhaltung gesteigert werden kann.

3.2. Prozessoptimierung bei der Mono-Vergärung von Energiepflanzen

Das technische Angebotspotenzial für eine Biogasnutzung aus Rückständen, Nebenprodukten und Abfällen ist immer noch stark ausbaufähig, aber auch begrenzt. Der zweite Teil der Arbeit konzentriert sich daher auf den Einsatz von speziell angebauten Energiepflanzen als Substrate für Biogasanlagen und den damit verbundenen verfahrenstechnischen Herausforderungen.

Der Einsatz von nachwachsenden Rohstoffen zur Biogaserzeugung ist jedoch im Vergleich zu Rückständen, Nebenprodukten und Abfällen mit höheren laufenden Kosten verbunden.

Abgeleitet von internationalen und nationalen Energie- und Klimaschutzzielen und der Notwendigkeit zur Stärkung der Eigenversorgung wurden nationale und regionale Regelungen und Unterstützungssysteme für den Ausbau Erneuerbarer Energie geschaffen. In Österreich ist für die Bereitstellung von Erneuerbarer Energie aus Biogas das Ökostromgesetz von besonderer Bedeutung. Die bundesweit einheitliche

Regelung trat 2002 in Kraft und enthielt außer den festgeschriebenen Einspeisetarifen keine Anforderungen, wie etwa den Gesamtwirkungsgrad der Anlagen (Ökostromgesetz, 2002). Die festgesetzten Tarife für Biogasstrom, aus landwirtschaftlichen Reststoffen oder Energiepflanzen führten in der Laufzeit dieser Regelung bis 31.12.2004 zu einem Genehmigungs- und späteren Ausbauboom landwirtschaftlich geprägter Biogasanlagen. Die eingesetzten Substrate bei diesen Anlagen sind mehrheitlich nachwachsende Rohstoffe. Diese werden landwirtschaftlich produziert und müssen von den Betreibern der jeweiligen Biogasanlagen extern zugekauft oder intern den Markpreisen entsprechend verrechnet werden.

Aus Sicht der definierten Zielsetzungen bedeutet dies, dass der Beitrag der Anaerobtechnologie zu energie- und umweltpolitischen Zielsetzungen in den Vordergrund tritt:

Wichtigstes Ziel ist es, möglichst viel Erneuerbare Energie bereit zu stellen. Dabei sollen die Produktionskosten des Biogases so gering wie möglich und der Beitrag zur Reduktion der Treibhausgasemissionen so groß wie möglich sein.

Die in den anderen Anwendungsfällen dominierenden Ziele der Abfallwirtschaft und des Umweltschutzes bekommen neue Schwerpunkte:

- Die möglichst hohe Reduktion der organischen Reststoffe wird durch die Notwendigkeit der höchstmöglichen Energieausbeute aus Energiepflanzen neu interpretiert.
- Umweltschutzziele, die bei der Abfallverwertung vor allem die Vermeidung der Emissionen in Luft und Gewässer und die Hygiene betreffen, werden um Fragen der nachhaltigen Produktion der Rohstoffe und der Ökobilanz des Gesamtsystems erweitert.

Die aus dem Zielsystem ableitbaren Herausforderungen für die Prozesskette Substratbereitstellung – Biogasproduktion – Gärrestverwertung und Gasnutzung sind in Abbildung 6 zusammengefasst.

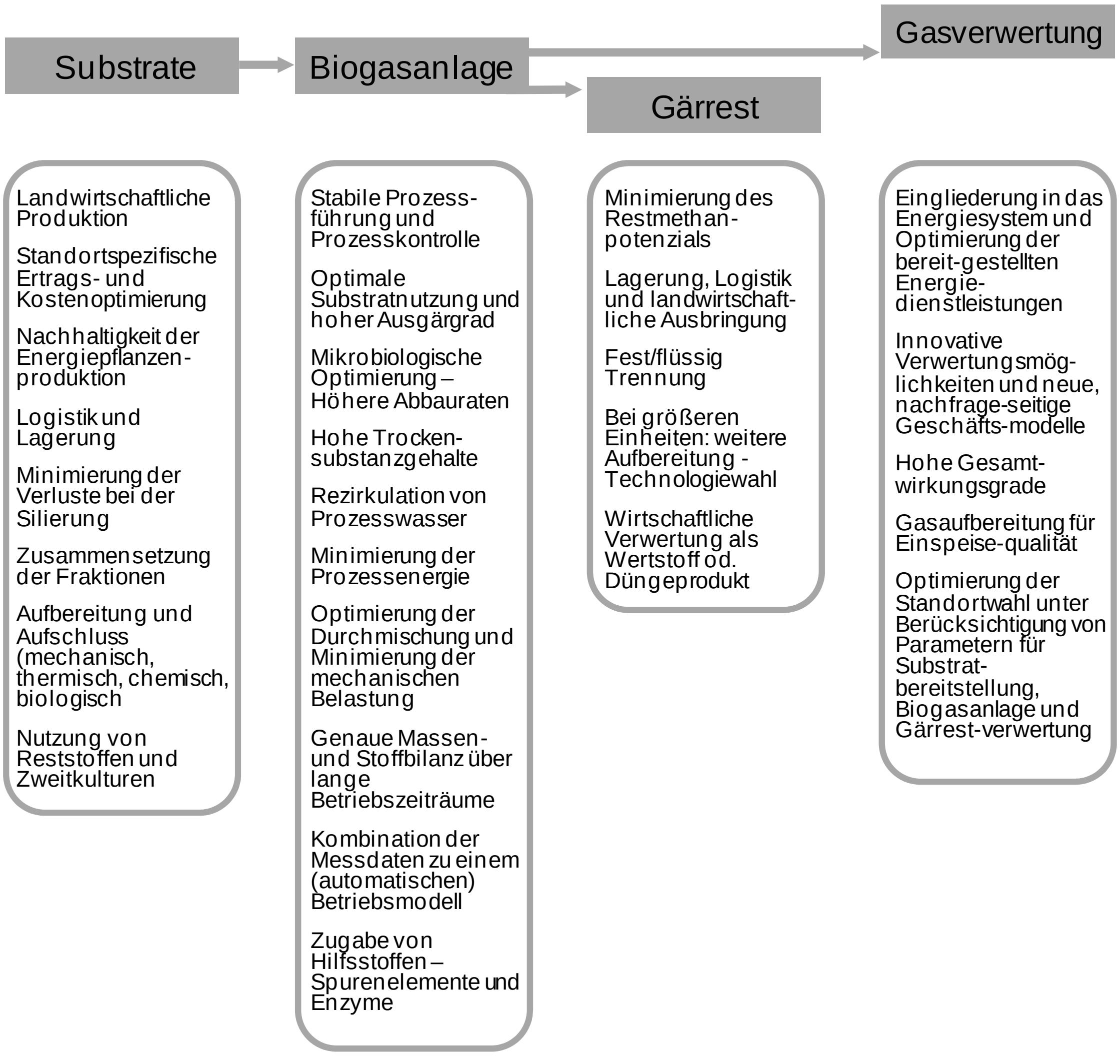

Abbildung 6: Herausforderungen bei der Biogaserzeugung aus Energiepflanzen

3.2.1. Produktion von Biogas aus nachwachsenden Rohstoffen

Die Erzielung einer möglichst hohen Energieausbeute aus den eingesetzten nachwachsenden Rohstoffen und die damit verbundenen verfahrenstechnischen Weiterentwicklungen sind nur bei Kenntnis der Stoff- und Energieströme in groß-technischen Anlagen möglich.

Über einen Zeitraum von 20 Monaten wurden daher die Massen- und Energieströme einer Biogasanlage auf Basis nachwachsender Rohstoffe ab der Inbetriebnahme im Februar 2005 bilanziert (Resch et al., 2008) [Publikation 2].

Diese Biogasanlage hat eine elektrische Leistung von 500 KW und war die erste Anlage in Österreich, die ausschließlich mit Energiepflanzen ohne Gülleeinsatz betrieben wird. Als Substrate werden Mais Ganzpflanzensilage sowie Gras und Kleemischungen eingesetzt. Eine Motivation für dieses Anlagenkonzept war durch den Strukturwandel in der Landwirtschaft nach dem EU Beitritt Österreichs 1995 gegeben, wodurch die Tier- und Weidehaltung im in der betrachteten exponierten Region vielfach aufgegeben wurde. Die Flächen, für die teilweise besondere Naturschutzauflagen gelten, müssen anderwärtig genutzt werden, um die Kulturlandschaft zu erhalten. Landwirtschaftliche Reststoffe wie Gülle stehen regional nur eingeschränkt zur Verfügung.

Daher sollte bei dieser Anlage aus ökonomischen und naturschutzrechtlichen Überlegungen ein Betrieb mit 100 % Gras und Klee möglich sein. Es wurde eine 2 stufige Biogasanlage mit volldurchmischten Rührkesseln errichtet, bei der die Substratzugabe über den Hauptfermenter erfolgt. Die organische Trockensubstanz der Substrate beträgt zwischen 30 % und 40 %. Die eingesetzten Gräser weisen einen hohen Faseranteil auf, der in der automatisierten verfahrenstechnischen Behandlung besonders herausfordernd ist. So wurden vom Anlagenhersteller langsam laufende querliegende Rührwerke entwickelt, die eine mechanische Durchmischung ermöglichen und in dieser Konfiguration erstmalig eingesetzt wurden.

Die Rührfähigkeit der Fermenterinhalte ist bis zu einem Trockensubstanzgehalt von maximal 10 % gewährleistet, der durch die Zugabe von Prozessflüssigkeit eingestellt werden muss. Da bei der untersuchten Anlage Gülle oder andere flüssige Fraktionen nicht zur Verfügung stehen, wird der Gärrest durch einen Schneckenseparator in eine feste und eine flüssige Phase getrennt. Aus der Menge des flüssigen Gärrests wird die notwendige Rezirkulationsflüssigkeit gewonnen. Diese wird kontinuierlich in den Hauptfermenter zugeführt.

Im Zeitraum der Inbetriebnahme und Bilanzierung lagen weder in Österreich noch in Deutschland zu dieser Betriebsweise Erfahrungen vor. Fischer und Krieg berichten 2006 von ersten errichteten Anlagen in Deutschland und skizzieren erste Erfahrungen. Auch sie weisen auf die Notwendigkeit der Rezirkulation von Prozessflüssigkeit und den in erster Linie mechanischen Herausforderungen hin.

3.2.2. Substratbereitstellung

Die Produktionskette der Substrate bis zur Lagerung im Silo hat einen wesentlichen Anteil an der Kostenstruktur und damit an der Wirtschaftlichkeit der Biogasanlage. Oftmals sind sie der größte Kostenfaktor. Volatile Preise haben daher einen direkten und wesentlichen Einfluss auf den erfolgreichen Anlagenbetrieb. Im Jahr 2007 betrug die durchschnittliche Einspeisevergütung für Biogasanlagen in Österreich 13,82 Cent kWh^{-1} (E-Control, 2009). Ein starker Anstieg der Rohstoffpreise von Juli 2007 bis Juni 2008 auf den Weltmärkten hatte Auswirkungen auf die lokalen Rohstoffpreise. Abbildung 7 zeigt die Preisentwicklung bei Mais im betrachteten Zeitraum. Obwohl die durchschnittliche Einspeisevergütung auf 17,71 Cent kWh^{-1} anstieg und damit zu einer breiten Diskussion über die Sinnhaftigkeit der Biogastechnologie führte, bedeuteten die Rohstoffpreise ernsthafte wirtschaftliche Probleme für viele Biogasanlagen. In der Novelle zum Ökostromgesetz im Jänner 2008 wurde daher die Möglichkeit des Preisausgleiches für Rohstoffe beschlossen und in der Tarifverordnung 2008 mit 4 Cent kWh^{-1} festgelegt. 198 Anlagen beantragten den Rohstoffzuschlag.

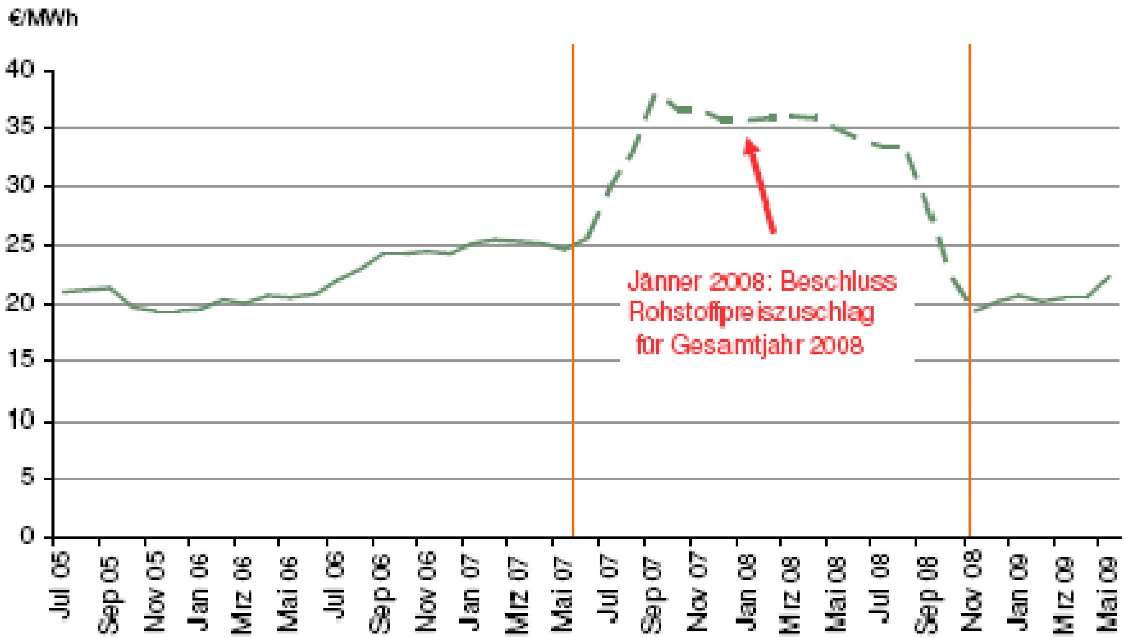

Abbildung 7: Preisentwicklung von Silomais (in Euro MWh-1 Brennstoffwärmeleistung) von Juli 2005 bis Mai 2009. (Quellen: Statistik Austria, Landwirtschaftskammer) In: E-Control (2009)

Dieses Beispiel unterstreicht, dass die Wirtschaftlichkeit einer auf Energiepflanzenbasis betriebenen Biogasanlage zu großen Teilen von den Produktionskosten der Energiepflanzen und der Effizienz der Energieausbeute beim anaerboben Prozess abhängig ist.

Die Pflanzenproduktion und neue Anbausysteme sind nicht Teil dieser Arbeit. Es liegen dazu umfassende nationale und internationale Untersuchungen vor (Karpenstein-Machan, 2001; Herrmann und Taube, 2006; Lehtomäki et al., 2008a; Prochnow et al., 2009; Bauer et al., 2010). Diese Beispiele legen dar, dass der Anbau von nachwachsenden Rohstoffen mit einem regional abgestimmten Konzept zu einer Erweiterung der Fruchtfolge führt.

Der wichtigste Parameter für die Auswahl von Energiepflanzen ist dabei der Nettoenergieertrag je Hektar. Unter österreichischen klimatischen Bedingungen und landwirtschaftlicher Infrastruktur haben Mais und mehrjährige Gräser das höchste Energiepotenzial (KTBL, 2006; Braun et al., 2009).

Durch eine Diversifizierung kann das jährliche Preis- und Ertragsrisiko des mehrheitlich eingesetzten Mais jedoch miniert werden (Vetter et al., 2008). Internationale und nationale Studien zeigen, dass durch die energetische Nutzung von Grünland ein geringeres Umweltrisiko als durch die intensive Maisbewirtschaftung besteht und durch Nutzung der Grünlandflächen die Kulturlandschaft und Biodiversität im Alpenraum erhalten werden kann (SRU, 2007; Buchgraber et al., 2010).

Bei der betrachteten Biogasanlage erfolgt die Produktion der Substrate im direkten Umkreis der Anlage. Mais und Grünlandpflanzen werden möglichst effizient geerntet und in Fahrsilos gelagert. Zur Logistik und zu Verlusten während der Silolagerung wurden Untersuchungen durchgeführt (Resch et al., 2007).

3.2.3. Prozessoptimierung der Energiepflanzenvergärung an Großanlagen

Die Biogasanlage wurde im Frühjahr 2005 nach einem selbst entwickelten und vorab definierten Plan hochgefahren. Die Dokumentation und daraus gezogene Schlüsse sind von Resch et al. (2005) zusammengefasst.

Die Darstellungen des Einflusses der Produktionskosten für Energiepflanzen verdeutlichen, wie wichtig es im laufenden Betrieb ist, einen möglichst hohen Ausgärgrad der eingesetzten Energiepflanzen zu erreichen.

Die wichtigsten Anforderungen an den Betrieb einer Energiepflanzenvergärung sind:

1. Volle Erfüllung des Einspeisevertrages nach dem Ökostromgesetz, da nicht gelieferte Mengen später nicht nachgeliefert werden können.

2. Kontrollierter mikrobiologischer Prozess und Stabilisierung des Trockensubstanzgehalts in beiden Fermentern

3. Höchstmögliche Methanausbeute und Energieumwandlung der eingebrachten Energiepflanzen, um die Kosten zu minieren.

Für einen wirtschaftlichen Betrieb sind alle drei Anforderungen zu erfüllen. Die limitierte Einspeisevergütung und die steigenden Substratpreise erfordern hocheffiziente Biogasanlagen.

3.2.4. Bilanzierung der Energiepflanzenvergärung ohne Güllezugabe in zwei Substratkombinationen

Die unter realen Bedingungen erzielbare Methanausbeute wurde in einer umfassenden Bilanzierung der Stoff- und Energieströme einer Biogasanlage ausgewertet und für zwei unterschiedliche Substratzusammensetzungen dargestellt (Resch et al., 2008) [Publikation 2]. Verglichen wird dabei die Variante 100 % Maiseinsatz mit Substratkombination von 52 % Gras und Klee und 48 % Mais.

Aus den Analysen der Massenflüsse und den Energiebilanzen kann geschlossen werden, dass ein Substratmix aus Mais, Klee und Gras die Notwendigkeit der Rezirkulation von flüssigem Gärrest erhöht, um einen maximalen Trockensubstanzgehalt von 10 % in den Fermentern nicht zu überschreiten. Ab dieser Größe steigt bei der betrachteten Anlage die Leistungsaufnahme der Rührwerke merklich an und das Risiko einer Betriebsstörung durch Schwimmschichten ist wahrscheinlich.

Im Zuge der Datenauswertung konnte die Leistungsaufnahme der Rührwerke als streng korrelierend mit dem Trockensubstanzgehalt im Bereich von 10 % TS ermittelt werden. Dieser, durch die Anlagensteuerung kontinuierlich erfasste Wert, dient nun bei der Großanlage als Regelgröße für den Substratinput und das Rezirkulationsmanagement.

Bei der Vergärung der eingesetzten Energiepflanzen ist die Hydrolyse der geschwindigkeitslimitierende Schritt in der Kette des anaeroben Abbaus. Materialien mit einem hohen Anteil an Lignocellulose werden nur langsam und unvollständig abgebaut, da der Ligninanteil anaerob nicht gespalten werden kann und somit die Struktur der Hemicellulose nur langsam aufgeschlossen werden kann (Scholwin et al., 2009).

Das eingesetzte Verhältnis der Substrate von 52 % Mais und 48 % Gras + Klee führt zu einer Verdoppelung der täglichen Rezirkulationsmenge und damit zu einer Reduktion der Aufenthaltszeit von jeweils 65 Tage auf 34 Tage. Die verkürzte Aufenthaltszeit und vor allem die erhöhte Ausschleusung von festem, noch nicht vollständig aufgeschlossenem, Gärrest lässt den Ausgärgrad der Energiepflanzen deutlich sinken. Das Restmethanpotenzial wurde regelmäßig in Batch-Ausgärversuchen bestimmt. Dabei wurden 70 % der in der Anlage nicht erschlossenen Substratenergie in der mit 7 % geringen Masse des festen Gärrests nachgewiesen. Aus Sicht des Betreibers steigt der zusätzlich notwendige Substratinput im Vergleich zu idealen Laborbedingungen in Batch Tests von 14 % auf 20 %. Dies bedeutet, dass um 6 % mehr Energiepflanzen zugeführt werden müssen, um die gesicherte Ökostrom Einspeisemenge ausnützen zu können.

Pfeifer (2007) bilanziert eine österreichische landwirtschaftliche Biogasanlage mit Gülleinput und ermittelt ein vergleichbares Restenergiepotenzial im Gärrest von 16,2 - 22,7 %. FNR (2010) berichtet im Messprogramm II für 61 Biogasanlagen in Deutschland von einem durchschnittlichen Restgärpotenzial von 91 Nm^3_{CH4} t_{OTS}^{-1} bei zweistufigen Anlagen bei einer Schwankungsbreite von 29 – 177 Nm^3_{CH4} t_{OTS}^{-1}. Bei der in dieser Arbeit betrachteten Anlage wurde das vergleichbare Restgärpotenzial im Endlager für die beiden Betriebsfälle Mais und Mais + Gras mit 50 bzw. 75 Nm^3_{CH4} t_{OTS}^{-1} ermittelt. Für den vollständigen Aufschluss dieser lignocellulose-haltigen Energiepflanzen sind daher lange Aufenthaltszeiten von mehreren Wochen oder Monaten notwendig, um eine vollständige Fermentation mit hohem Gasertrag und minimalem Restmethanpotential zu erreichen (Weiland, 2010). Lethomäki et al. (2008), Nordberg et al. (2007) sowie Kaparaju und Rintala (2008) untersuchen im Labormaßstab die Vergärung von nordischen, grasartigen Energiepflanzen und landwirtschaftlichen Reststoffen in Rezirkulationssystemen.

Wichern et al. (2009) modellieren auf Basis vorliegender Versuchsergebnisse die Energiebilanz der Pflanzenvergärung in einer Weiterentwicklung des Anaerobic Digestion Model No. 1 und bauen dabei auf den Publikationen von Lindorfer et al. (2007) [Publikation 5] und Resch et al. (2008) [Publikation 2] auf. Sie entwickeln die notwendigen Modellparameter für lignozellulosereiche Energiepflanzen aus Laborversuchen und theoretischen Überlegungen.

Der niedrigere und stabilere Preis der eingesetzten Gras- und Kleesilagen und die Neuheit der Technologie rechtfertigten in der Vergangenheit die noch reduziertere Methanausbeute. Langfristig muss aber die Energieausbeute bei diesen Energiepflanzen weiter gesteigert werden.

Eine Möglichkeit dazu ist die Vorbehandlung der Substrate mit chemischen, physikalischen und thermischen Verfahren. Ziel dieser Verfahren ist es, die Stärke und Zellulose verfügbar zu machen, indem der lignifizierte Verband aufgebrochen wird. Bei Systemen wie am Beispiel der diskutierten Biogasanlage könnte es auch sinnvoll sein, den abgepressten festen Gärrest weiter zu behandeln. Die feste Phase entspricht etwa nur 10 % der Gärrestmasse und enthält nahezu alle fasrigen Bestandteile, die durch eine weitere Behandlung anaerob verfügbar gemacht werden können. Perez-Lopez et al. (2005) führte mit dieser Fraktion erste Laboruntersuchungen durch, die zu einem Verfahren weiter entwickelt werden könnten. Vergleichbare Ansätze wurden von Jagadabhi et al. (2008) publiziert.

Bauer et al. (2009) beschreiben die Vorbehandlung von lignozellulosehaltigem Stroh mit Steam-Explosion. Bei diesem thermischen Aufschlussverfahren wird das Stroh gemeinsam mit Prozesswasser unter Druck erhitzt und spontan entspannt. Dadurch soll der Ligninverband aufgebrochen werden. In die erfolgreichen Vergärungsversuche ist keine Betrachtung der eingesetzten Energie für dieses Verfahren inkludiert, die für die Bewertung des Nettoenergieertrags aber notwendig ist.

3.2.5. Prozessbeeinflussung durch die Steigerung der Energiepflanzenzugabe

Die definierten Anforderungen an den wirtschaftlichen Betrieb einer Energiepflanzenvergärung, wie die vollständige Erfüllung des Einspeisevertrags für Ökostrom und die möglichst hohe Nutzung der Substratenergie, gelten auch bei der Steigerung der Zugabe von Energiepflanzen zu einer bestehenden Anlageninfrastruktur. Lindorfer et al. (2007) [Publikation 5] untersuchten den Einfluss der Leistungssteigerung durch zusätzliche Energiepflanzenzugabe bei einer mit Gülle und Energiepflanzen betriebenen landwirtschaftlichen Biogasanlage. Der zusätzliche Einsatz der Substrate bei existierender Infrastruktur bedeutet somit eine Erhöhung der organischen Raumbelastung und eine Verkürzung der Aufenthaltszeit. Die Untersuchungen an dieser Anlage zeigten, dass eine deutliche Steigerung der Biogasproduktion durch den Energiepflanzeneinsatz erreicht werden konnte. Negative Konsequenz war die Zunahme des Restmethanpotentials im Gärrest. Als Konsequenz der Messungen und der energetischen und ökonomischen Bilanzierung wurde das Endlager der

Großanlage abgedeckt. Lindorfer et al. (2008) zeigen in weiterführenden Untersuchungen, dass auch bei hoher Raumbelastung ein instabiler Prozess durch intensive Prozesskontrolle sicher gestellt werden kann.

3.2.6. Auswirkungen der Energiepflanzenvergärung auf die Umweltschutzziele

Die beschriebene Nutzung von nachwachsenden Rohstoffen zur Biogas- und anschließenden Stromproduktion ist mit Umweltauswirkungen verbunden, die je nach implementierter Prozesskette stark schwanken können. Zusätzlich sind bei der Verwendung von Pflanzen zur Energieproduktion Zielkonflikte mit Nahrungs- und Futtermittelproduktion gegeben. Im Vergleich zu stofflichen Nutzung von Biomasse werden Konkurrenzen in der Prozesskette aufgezeigt.

Die nachhaltige Produktion der Energiepflanzen für alle Formen der Bioenergie und die Ökobilanz der Prozesse rücken in den Mittelpunkt der Umweltschutzziele. Sie bilden den argumentativen Gegenpart zur Steigerung der Erneuerbaren Energie und den damit verbundenen Zielen. Die Reduktion der Treibhausgasemissionen und die Berechnung von Ökobilanzen sind Teil der Umweltziele und das verbindende Thema zur Energieproduktion. Die Netto-Reduktion der Treibhausgasemissionen durch die betrachteten Technologien und Prozessketten ist daher eine anerkannte Vergleichsgröße.

Eine Ökobilanzierung oder Berechnung der Treibhausgasreduktion wurde für die diskutierten Anlagenbeispiele nicht durchgeführt. In der Literatur wurden - aus sehr unterschiedlichen Motivationen – Bilanzierungen für Bioenergiesysteme und auch den Biogasprozess publiziert.

Berglund und Börjesson (2006), Börjesson und Berglund (2006) sowie Börjesson und Berglund (2007) analysieren die Biogasproduktion aus Energiepflanzen und landwirtschaftlichen Reststoffen unter schwedischen Bedingungen. Wie im dargestellten Beispiel der Energiepflanzenvergärung identifizieren auch sie den größten Einfluss durch die Bereitstellung, die Transportwege und die Qualität der Substrate, die Energieeffizienz der Biogasanlage und die Gasnutzung. Als die größten Faktoren eines effizienten Gesamtsystems sehen sie die Substratbereitstellung und Restmethan-Emissionen im Gärrest.

Eine kritische Bilanz der Biogasproduktion aus Mais ziehen Herrmann und Taube (2006). Aufgezeigt werden unter anderem die Substratproduktion und Logistik der intensiven Maiskulturen sowie die negativen Auswirkungen der Mais-Monokulturen auf die Biodiversität und Landnutzung. Salter and Banks (2009) bilanzieren das System Biogas nach Energieströmen und kommen zu dem Schluss, dass die Produktion der Pflanzen und der Düngereinsatz die meisten Ressourcen konsumieren und daher auf ein vollständiges Gärrestmanagement zu achten ist. Braun und Laaber (2007) stellen das Input:Output Verhältnis der Energiemengen bei vier typischen österreichischen Biogasanlagen dar. Die Resultate zeigen, dass eine Erhöhung des Anteils an Gülle und Wirtschaftsdünger die Bilanz ebenso deutlich verbessert wie die optimale Nutzung der Abwärme des BHKWs.

Das Umweltbundesamt bilanziert im Klimaschutzbericht 2010, dass die energetische Nutzung der Wirtschaftsdünger in Biogasanlagen eine effiziente Klimaschutzmaßnahme mit doppeltem Nutzen ist: Methanemissionen aus dem Wirtschaftsdüngermanagement werden vermieden – bei gleichzeitiger Gewinnung Erneuerbarer Energie. Aufgrund des gültigen Ökostromgesetzes besteht allerdings ein Anreiz zur Vergärung von Energiepflanzen. Dadurch ist die Nutzung von Wirtschaftsdüngern in Biogasanlagen geschmälert und Emissionsreduktionen werden nicht realisiert.

Diese und andere Bilanzierungen des Nettobeitrags zur Steigerung Erneuerbarer Energie oder zur Treibhausgasreduktion machen deutlich, wie wichtig ein effizienter Prozess ist, der die Produktion und Bereitstellung der Energiepflanzen, den mikrobiologischen und verfahrenstechnischen Prozess in der Biogasanlage, die sinnvolle Verwertung des Gärrests und die Nutzung des Gases mit einem höchstmöglichen Beitrag zur Nutzenergie beinhaltet. Die dargestellten Analysen an den ersten in Österreich errichteten Großanlagen zeigen auf, dass eine energieeffiziente Produktion Erneuerbarer Energie möglich ist und erreicht wird. Es besteht aber weiter dringender Verbesserungsbedarf, der durch neue Entwicklungen in der Anlagentechnik sowohl durch wissenschaftliche Forschung im angewandten als auch im Grundlagenbereich erreicht werden muss.

4. Schlussfolgerungen

Die Erzeugung von Biogas ist eine wesentliche Komponente des schonenden Umgangs mit Ressourcen. Der vielfältige Einsatz der Anaerobtechnologie leistet damit wichtige Beiträge zur Erfüllung von Vorgaben im Bereich des Umweltschutzes und der Abfallwirtschaft. In vielen Fällen wird durch das produzierte Biogas Erneuerbare Energie bereit gestellt und damit die durch internationale Energie- und Klimaziele steigende Nachfrage nach Erneuerbarer Endenergie bedient. Dies gilt sowohl für die Verwertung von Reststoffen und Rückgewinnung von Wertstoffen als auch für die Nutzung von Energiepflanzen.

Es sind allerdings in beiden Anwendungsgebieten noch bedeutende Optimierungsschritte möglich, um einen effizienteren Abbau der Substrate oder eine Steigerung in der Energieumwandlung zu erzielen. Für ausgewählte Herausforderungen in diesen Bereichen werden in dieser Arbeit und den inkludierten Publikationen Lösungsansätze in Form von Verfahrensvorschlägen oder Bilanzierungen präsentiert.

Verfahrensoptimierung bei problematischen Reststoffen

Durch die Konzentration zu größeren Produktionsstandorten in der Lebensmittelindustrie können etablierte regionale Stoffkreisläufe nicht mehr genutzt werden. Obwohl die Produktion selbst effizienter wird fallen mehr Reststoffe und Nebenprodukte an, für die ökonomisch und ökologisch sinnvolle Verwertungswege gefunden werden müssen. Am Beispiel eines Schlachthofes wurde gezeigt, dass eine Biogasanlage umfassend in den Produktionsbetrieb integriert werden kann und somit Stoff- und Energiekreisläufe geschlossen werden können.

Teile der anfallenden tierischen Nebenprodukte werden in diesem Energiesystem zu Biogas und in weiterer Folge in einem BHKW zu Ökostrom umgewandelt. Der mikrobiologische Vergärungsprozess ist durch den hohen Proteingehalt der tierischen Nebenprodukte und damit durch die hohe Ammonium- bzw. Ammonikakonzentration bei der Monovergärung in den Fermenterinhalten gehemmt. Durch langjähriges Monitoring und intensive Prozesskontrolle konnte der Vergärungsprozess stabilisiert werden. Eine dauerhafte Steigerung der Zugabe von noch zu Verfügung stehenden Nebenprodukten, vor allem Blut, ist im bestehenden Anlagensystem nicht möglich.

Die Reduktion des Stickstoffgehaltes in den Fermentern sollte zu einer Verbesserung der Abbauleistung führen und die Verwertung zusätzlicher Substratströme ermöglichen. Es wird daher ein Verfahren vorgeschlagen, mit dem das Ammonium direkt aus dem Prozess entfernt werden soll. Die Funktionsweise der ersten beiden Verfahrensschritte Desaminierung und Strippung des Ammoniaks wurden in Vorversuchen getestet. Die positiven Auswirkungen der Stickstoffreduktion auf den Vergärungsprozess wurden in kontinuierlichen Versuchen nachgewiesen. Dabei wurden mehrere zukünftige Betriebszustände der Großanlage simuliert und wahrscheinliche Grenzen für eine Kapazitätserweiterung aufgezeigt.

Aus den präsentierten Versuchsergebnissen lässt sich ableiten, dass durch eine teilweise Entfernung des Stickstoffs eine deutliche Steigerung der Abbauleistung und eine Stabilisierung der Fermentation erreicht werden. In einem nächsten Entwicklungsschritt sollte die Stickstoffentfernung im Pilotmaßstab in die Großanlage integriert und weiter entwickelt werden.

Durch die Verwertung von tierischen Nebenprodukten in standortintegrierten Biogasanlagen und die Verwertung des Gärrests als Dünger wird der Stickstoffkreislauf geschlossen. Wird zusätzlich durch die Entfernung und Rückgewinnung von Ammonium ein definierter flüssiger Dünger bereit gestellt, können energieintensiv produzierte Mineraldünger weitgehend ersetzt werden.

Der beschriebene Verfahrensweg erfüllt abfallwirtschaftliche Zielsetzungen durch die Verwertung organischer Reststoffe und die Rückführung von Wertstoffen in den Düngekreislauf. Dadurch kann der Einsatz fossiler Energie bei der Produktion von Mineraldünger substituiert werden.

An diesem Anlagenstandort wurde in weiteren Arbeiten eine Integration der Energieverwertung des Biogases in den Produktionsprozess der Anlage erreicht. Durch die Installation eines Wärmespeichers konnte das gleichmäßige Abwärmeangebot des BHKW mit der produktionsbedingten Nachfrage in Übereinstimmung gebracht werden. Dieses integrierte Energiesystem ist ein erfolgreiches Beispiel, wie die Erfüllung von Vorgaben des Umweltschutzes mit der Produktion Erneuerbarer Energie und der direkten Substitution fossiler Energieträger in Einklang gebracht werden können.

Bilanzierung der Biogasproduktion aus Energiepflanzen

Einen neues Einsatzgebiet der Anaerobtechnologie ist die Produktion Erneuerbarer Energie aus nachwachsenden Rohstoffen. Besonders in Österreich und Deutschland wurden in den letzten Jahren durch Einspeisetarife für Ökostrom Rahmenbedingungen geschaffen, die diese Anlagensysteme forcieren.

Für eine Weiterentwicklung der Verfahren ist es sinnvoll, bestehende Großanlagen genau zu bilanzieren und daraus Schlüsse zu ziehen. Die wichtigsten Ziele beim Betrieb dieser Anlagen müssen sein, durch einen nahezu unterbrechungsfreien Betrieb den befristeten Einspeisevertrag bestmöglich zu erfüllen und bei den eingesetzten Substraten einen möglichst hohen Ausgärgrad zu erreichen. Beides stärkt die Wirtschaftlichkeit der Anlagen, da einerseits die Ertragsseite durch den Verkauf des Ökostroms maximiert wird und andererseits durch die höchstmögliche Nutzung der Energie die Substratkosten minimiert werden.

Im Rahmen der durchgeführten Projekte wurde bei einer Biogasanlage, die ausschließlich mit nachwachsenden Rohstoffen betrieben wird, eine Stoffstrom- und Energiebilanz erarbeitet. Es wurde dabei die Verwendung von zwei Substrat-kombinationen untersucht. Verglichen wurde die Verwendung von Mais Ganzpflanzen-silage mit einer Kombination aus Mais und Klee- + Grasmischungen. Diese Grünland-pflanzen können am Anlagenstandort in extensiver Bewirtschaftung und unter Naturschutzauflagen bereit gestellt werden.

In der Bilanzierung einer Großanlage konnte gezeigt werden, dass eine Vergärung von hohen Gras- und Kleeanteilen möglich ist. Der höhere Faseranteil dieser Substrate macht eine höhere Flüssigkeitszugabe notwendig, um einen Trockensubstanzgehalt in den Fermentern einzustellen, der eine weitere Rührfähigkeit sicher stellt. Die Prozessflüssigkeit wird durch das Abpressen des Gärrests in eine feste und flüssige Phase gewonnen und in den ersten Fermentationsschritt rezirkuliert. Die Steigerung dieses Rezirkulationsstroms beim Einsatz von Gras und Klee führt zu einer reduzierten Methanausbeute, da die Aufenthaltszeit reduziert wird und das Energiepotenzial des festen Gärrests nicht weiter genutzt werden kann.

Trotz der in dieser ersten Bilanzierung festgestellten reduzierten Ausbeute – und des damit erforderlichen Mehrbedarfs zur Auslastung der Anlage – hat die Verwendung von Gras und Klee neben ökologischen Gründen auch ökonomische Vorteile. Anders als bei Mais ist das Preisrisiko für diese Grünlandsubstrate geringer und ein stabiler, regionaler Preis kann zwischen Rohstoffproduzenten und Anlagenbetreibern leichter gefunden werden.

Beim weiteren Ausbau der Nutzung von Energiepflanzen zur Biogasproduktion muss die maximale Ausnutzung der Substratenergie erreicht werden. Weiters muss der Energiegehalt des Biogases mit einem hohen Gesamtwirkungsgrad genutzt werden. Bei der Ökostromproduktion in BHKWs bedeutet dies durch Standortwahl und System-auslegung einen großen Teil der Wärme in sinnvollen Anwendungen zu nutzen. Bei einer Aufbereitung des Gases zu Biomethan und der Substitution von Erdgas müssen effiziente Aufbereitungstechnologien eingesetzt werden. Bei der Verwendung des Gases in Kraft – Wärme – Kopplungen, als Treibstoff oder in der industriellen Produktion muss ein hoher Wirkungsgrad bei der Gasverwendung sicher gestellt wird.

Bei erfolgreicher Umsetzung dieser Ziele werden der Nutzung von nachwachsenden Rohstoffen zur Biogasproduktion auch in kritischen Ökobilanzen und Lebenszyklus-analysen geringere Umweltauswirkungen als anderen Bioenergietechnologien beschieden (Kaltschmitt und Streicher, 2009).

Ausblick

Die Erkenntnisse aus den durchgeführten Projekten zeigen, dass bei der Weiter-entwicklung der Anaerobtechnologie eine noch stärkere Verbindung der Grundlagen-forschung zu mikrobiotechnologischen Fragestellungen mit den Herausforderungen an Großanlagen geschaffen werden muss. Die Ausrichtung auf gegebene oder zukünftige Ziele im Umweltschutz, in der Abfallwirtschaft, bei Erneuerbaren Energien oder der Reduktion von Treibhausgasemissionen muss dabei frühzeitig gegeben sein.

Bei der Nutzung von Biomethan als Quelle Erneuerbarer Energie wird es in Zukunft wichtig sein, eine nachfrageorientierte Herangehensweise zu finden. Die Integration in industriellen Anwendungen ist eine bereits etablierte Möglichkeit. Ein weiterer Schritt könnte die Umsetzung von Projekten sein, die Biomethan als alternatives Ökogas vermarkten und die vorhandene Infrastruktur und Anwendungstechnologien für Erdgas nutzen.

Dafür sollten neue Rahmenbedingungen entwickelt und implementiert werden. Die potenzielle Nachfrage und die vorhandene Netzinfrastruktur in Ballungsräumen bieten sich für neue (Öko)Gasprodukte im Strom, Wärme und Treibstoffmarkt besonders an. Erste Projekte, getragen auch von etablierten Gasversorgern, sind auch in Österreich in Umsetzung und tragen auch dazu bei, den Stellenwert der Technologie in der Gesellschaft zu steigern. Ein europaweites Nachweissystem für diese Produkte in Reinverwendung oder auch in Mixprodukten würde das Kundenvertrauen stärken.

5. Referenzen

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BHKW	Blockheizkraftwerk
BMLFUW	Bundesministerium für Land- und Forstwirtschaft, Umwelt und Wasserwirtschaft
BMWFJ	Bundesministerium für Wirtschaft, Familie und Jugend
BREF Document	Best Available Techniques Reference Document
BSE	*Bovine spongiforme Enzephalopathie*
CH_4	Methan
CO_2	Kohlendioxid
COD	Chemical Oxygen demand (entspr. CSB: Chemischer Sauerstoffbedarf)
E-Control	Energie-Control GmbH
EEG	Erneuerbares Energien Gesetz (Deutschland)
EU	Europäische Union
$g\ kg^{-1}$	Stoffkonzentration in Gramm pro Kilogramm
$g\ l^{-1}$	Stoffkonzentration in Gramm pro Liter
GWh	Gigawattstunden
IEA	International Energy Agency
$kg_{COD}\ m^3\ d^{-1}$	Organische Raumbelastung in kg COD je m^3 Fermentervolumen und Tag
kWel	Elektrische Engpassleistung in Kilowatt
MW	Megawatt
OeMAG	Abwicklungsstelle für Ökostrom AG
TNP	Tierische Nebenprodukte
USGS	US Geological Service

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3. Enhancement options for the utilization of nitrogen rich animal by-products in anaerobic digestion

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4. Anaerobic degradation of animal by-products

Roland Kirchmayr, Christoph Resch, Martin Mayer, Stefan Prechtl, Martin Faulstich, Rudolf Braun, Johann Wimmer

in: Oreopoulou, V., Russ, W. (Eds.), Utilization of By-Products and Treatment of Waste in the Food Industry, Springer, New York, pp.159-191.

5. **The impact of increasing energy crop addition on process performance and residual methane potential in anaerobic digestion**

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Optimised anaerobic treatment of house-sorted biodegradable waste and slaughterhouse waste in a high loaded half technical scale digester

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Abstract Anaerobic co-digestion of organic wastes from households, slaughterhouses and meat processing industries was optimised in a half technical scale plant. The plant was operated for 130 days using two different substrates under organic loading rates of 10 and 12 $kg_{COD}.m^{-3}.d^{-1}$. Since the substrates were rich in fat and protein components (TKN: 12 $g.kg^{-1}$) the treatment was challenging. The process was monitored on-line and in the laboratory. It was demonstrated that an intensive and stable co-digestion of partly hydrolysed organic waste and protein rich slaughterhouse waste can be achieved in the balance of inconsistent pH and buffering NH_4-N. In the first experimental period the reduction of the substrate COD was almost complete in an overall stable process (COD reduction $>82\%$). In the second period methane productivity increased, but certain intermediate products accumulated constantly. Process design options for a second digestion phase for advanced degradation were investigated. Potential causes for slow and reduced propionic and valeric acid degradation were assessed. Recommendations for full-scale process implementation can be made from the experimental results reported. The highly loaded and stable co-digestion of these substrates may be a good technical and economic treatment alternative.

Keywords Anaerobic digestion; half technical scale; high nitrogen concentrations; slaughterhouse wastes

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Introduction

Anaerobic biological degradation processes are an ideal treatment technology for organic wastes. However, prior to the application of new waste streams as substrates, series of investigations are required to develop and optimise efficient and stable treatment processes (Verstraete *et al.*, 2004). Various concepts for the treatment of the organic fraction of municipal solid wastes (OFMSW) are available (Bolzonella *et al.*, 2003; Luning *et al.*, 2003). This work is based on a pre-treatment concept that has been installed at the site of the project partner SESA in Este/Padova (I) in which the OFMSW is pressed into a liquid and a solid part. The solid fraction undergoes aerobic composting and the liquid fraction is highly suitable for anaerobic digestion (Held *et al.*, 2001). The main objective of the project reported here was to assess the feasibility of co-digestion of the pressed liquid of OFMSW and waste streams from slaughterhouses and the meat processing industry. Due to changes in the market and in regulation (e.g. Animal by-product regulation (ABP) (EC) No 1774/2002), slaughterhouse waste is a new challenge to be treated anaerobically (Kirchmayr *et al.*, 2003). A stable anaerobic degradation process under very high organic and nitrogen loading rates should be established. Since the free ammonia concentration has been suggested to be the active component which causes ammonia inhibition (Angelidaki and Ahring, 1993), the investigations focused on behaviour of the degradation process under high nitrogen loads. Secondly, as for highly biodegradable wastes containing fats it is advisable to use two phase anaerobic digestion processes (Mata-Alvarez *et al.*, 2000; Peirera *et al.*, 2002): the first phase with a high organic load was investigated in a

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$4\,m^3$ half technical scale plant; and to assess the feasibility of an entirely treatment concept at full technical scale, a second digester was employed to investigate the degradation process in batch experiments.

Material and methods

The half technical scale plant was set up at the site of SESA. The plant consisted of a $0.72\,m^3$ hygienisation unit and a $4\,m^3$ completely stirred anaerobic digester (Figure 1). The system was fully controlled by a programmable-logical-controller and equipped with the following on-line measuring devices: the hygienisation unit with temperature sensor and pressure control for level management; the anaerobic digestion unit with temperature control, gas pressure control and gas flow meters. The gas composition (CH_4, CO_2, H_2, and H_2S) was monitored using an industrial scale on-line gas analyser by AWITE (D). The chemical parameters COD, NH_4-N, TKN and volatile fatty acids (VFA) were analysed according to standard methods. All batch experiments for methane yield determination were carried out as described by Grasmug (2004). The hygienisation unit was operated as stipulated in the EU ABP directive for category III material: particle size < 12 mm; $T > 70\,°C$ for $t > 60$ min. The anaerobic digester was inoculated with active biomass from an existing full scale plant treating similar substrates. It was operated under mesophilic conditions (37.5 °C) and a HRT of 21 days. The substrates were sourced from the SESA organic waste treatment process.

The OFMSW in the Veneto region are collected door-to-door and consist therefore only of kitchen waste and (industrial) food processing residues. The separated liquid fraction is highly suitable for anaerobic digestion (TSS: 17%; VSS: 13 %). The potential methane yield of several additional wastes was tested in batch experiments. Of the wastes tested, fractions taken from poultry slaughterhouses (blood, fat, carcasses) and meat processing industry (concentrated animal fat and proteins) were chosen for half technical scale experiments. Two different substrate mixtures from these waste streams were made based on the pressed liquid of OFMSW. For experiment periods 1 and 2 the substrates were composed as listed in Table 1. An organic loading rate of $10\,kg_{COD}.m^{-3}.d^{-1}$ was appointed in period 1 and the substrate was constantly fed every 3 hours for 67 days. In experimental period 2 the substrate was newly composed and the organic loading rate was set to $12\,kg_{COD}.m^{-3}.d^{-1}$. The hydraulic retention time was kept constant at 21 days.

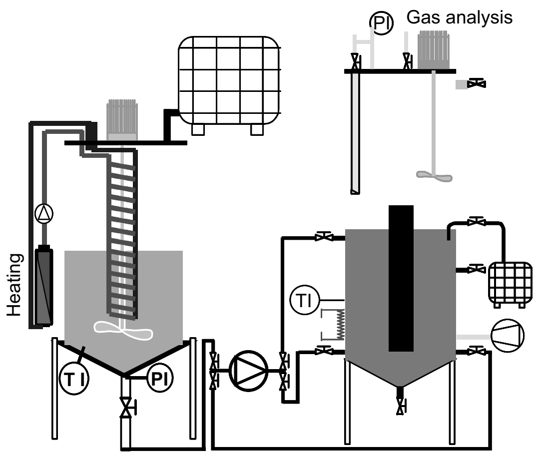

Figure 1 Half technical scale plant including a $0.7\,m^3$ hygienisation unit and a $4\,m^3$ anaerobic digester and various pumps and control devices

Table 1 Characteristics of composed substrates for experiment periods 1 and 2

	Substrate mix period 1 50% poultry blood and fat 50% pressed liquid of OFMSW	Substrate mix period 2 30% concentrated animal proteins and fat 70% pressed liquid of OFMSW
TSS ($g.kg^{-1}$)	113.5	198.1
VSS ($g.kg^{-1}$)	89.0	161.2
VSS/TSS (%)	81.5	79.3
COD ($g.kg^{-1}$)	263.20	330.10
TKN ($g.kg^{-1}$)	8.56	12.17

To show the technical or economical necessity of a second fermentation phase, the effluent of the pilot plant was further treated in laboratory batch experiments. One effluent sample from experimental period 1 (1) and two samples from period 2 (2/1 and 2/2) were collected. The methane yield, COD degradation and the development of VFA concentration were investigated over 99 days.

Results and discussion

The substrate mixtures were composed to optimise the methanization process for the pressed liquid of OFMSW and to use synergetic effects of inconsistent pH and buffering NH_4-N to avert process inhibitions (Angelidaki *et al.*, 1993; Speece, 1996). Advantages and disadvantages of the applied waste streams in mono-substrate digestion are documented in the literature (Mata-Alvarez *et al.*, 2000; Salminen *et al.*, 2000) and are well known in practice. These are outlined in Table 2.

As the inoculum for the half technical scale experiments was taken from the AD plant operating at SESA, no start up phase was needed and the full organic load ($10\,kg_{COD}.m^{-3}.d^{-1}$) could be added from the first day on. The gas production started immediately, with a content of between 65–70% methane (average: 68%) caused by the high protein content of the substrate (Salminen *et al.*, 2000). The methane productivity was stable over the whole experiment period at $2.22\,Nm^3_{CH4}.m^{-3}_{Reactor}.d^{-1}$. All measured parameters demonstrated that the digestion process was effective and stable. Characteristic metabolites such as volatile fatty acids did not reach inhibitory levels (Figure 2a). The COD degradation rate was stable around 80 % and the methane yield $60\,Nm^3_{CH4}.Mg^{-1}_{Substrate}$. Even intended shock loads did not destabilise the process (detailed data not shown). This underlines the fact that the microbiological degradation process was definitely not overloaded. This became apparent when the feed was stopped at day 67 and the VFA concentrations decreased rapidly within 13 days. Results for period 1 are displayed in Figure 2. The high nitrogen content of the substrate did not seriously inhibit the process either. 80% of the TKN was converted into soluble NH_4-N. As the pH remained stable and did not exceed 8.0, the dissociation equilibrium did not change towards toxic unionized

Table 2 Advantages and disadvantages of organic waste in mono-substrate digestion processes

	Advantages	Disadvantages
Pressed liquid of OFMSW	Easily available for micro-organisms because of commenced hydrolyses; buffered and low nitrogen content	Medium gas yield Contraries as plastics and sand
Poultry slaughterhouse waste	High methane yield	High nitrogen content, NH_4-N buffered; foaming during hygienisation
Concentrated animal proteins and fat	High methane yield	High nitrogen content and slow degradable fats (LCFA)

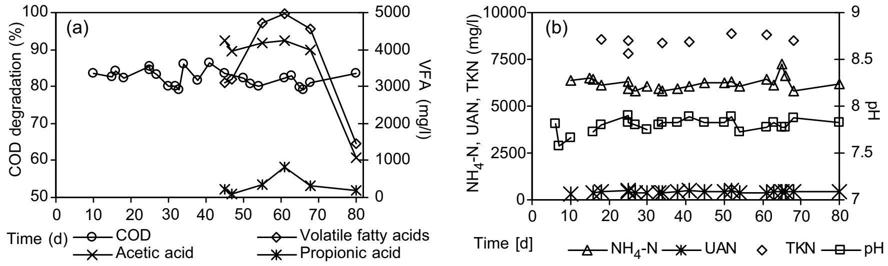

Figure 2 a) COD degradation rate and volatile fatty acid concentrations b) Nitrogen related parameters (TKN, NH_4-N, UAN, pH) in experiment period 1

ammonia (UAN). The UAN concentration had a maximum concentration of 500 mg.kg^{-1}, well below the critical level of 1,000 mg.kg^{-1} (Hansen *et al.*, 1998).

In experimental period 2 (results shown in Figure 3) the substrate mix was changed (Table 1) and the organic loading rate was increased to over 12 kg$_{COD}$.m^{-3}.d^{-1} to investigate the operating limits of the system. The TKN concentration in the digester increased to 11.85 g.kg^{-1}. This resulted in an NH_4-N concentration of 10 g.kg^{-1} and a UAN concentration of up to 1,200 mg.l^{-1}. An indicator of a strongly inhibited or severely imbalanced process is a rapid reduction in methane production. This reduction was not observed since the methane productivity was very high (3.15 $Nm^3_{CH4}.m^{-3}_{Reactor}.d^{-1}$) over the whole feeding period of 48 days. The gas produced contained 67–72% methane. Through on-line or on site measurement no possible inhibition or instability was evident. Further investigations showed the following process development: the COD degradation rate decreased from 80% in the first 25 days down to 68% at the end of the experiments. Concurrently, the methane yield changed from 102 to 94 $Nm^3_{CH4}.Mg^{-1}_{Substrate}$. Substrate components and metabolites that were less degradable accumulated in the digester and in the residues. The volatile fatty acid concentrations, especially propionic acid, increased. After the substrate feed was stopped on day 48 the system was investigated for a further 2 weeks without additional feed: the acetic acid concentration decreased rapidly by 85%, whereas the propionic acid was not further degraded.

Additional batch experiments to assess the feasibility of a second digestion phase

The degree of decomposition that could be achieved in 21 days HRT in the half technical scale pilot plant was not satisfactory for direct use of the effluent as a quality end product. The potential for further degradation of metabolites like long chain and volatile fatty acids was investigated in laboratory batch experiments. These tests should evaluate

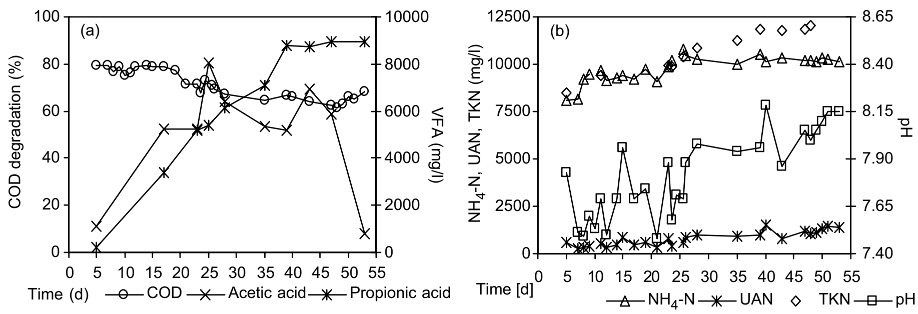

Figure 3 a) COD degradation rate and volatile fatty acid concentrations. b) Nitrogen related parameters (TKN, NH_4-N, UAN, pH) in experimental period 2

the necessity of a second digestion phase to obtain a higher methane yield and a better quality end product. Therefore, samples of the pilot plant effluent from experimental periods 1 and 2 were taken. The sample from period 1 (1) was taken on day 67 when the continuous feeding was stopped. The first sample from period 2 (2/1) was taken on day 24 and the second sample (2/2) on day 48 (the last day of feeding). The methane yields based on Mg effluent are charted in Figure 4a. Nevertheless the effluent from experimental period 1 was already degraded by 82%, an additional COD reduction of 5.4 percentage points out of total COD could be achieved.

The effluents from experiment period 2 gave different results. The methane production of sample 2/1 started immediately, whereas a low productive lag-phase was observed for test 2/2. Sample 2/2 produced more methane than the other samples, because of the highest residual COD in effluent 2/2. Though Figure 4a shows the highest methane yield for sample 2/2, the yield per kg COD was approximately the same for sample 2/1 and 2/2 ($140\,Nm^3.kg^{-1}_{COD}$).

Far more important than the total additional methane yield over a longer testing, is the accessory achievable methane production and COD reduction within a technically feasible and economical time period. To assess the options for a second digestion step, the methane productivity was categorised in days of digestion for each sample (Figure 4b). Sample 1 had a considerable methane productivity of $0.17\,Nm^3.m^{-3}.d^{-1}$ within the first 10 days of retention time. A small second fermentation phase guaranteeing these additional 10 days of digestion will be enough to remove most of the residual COD from the easy available substrate mixture of pressed liquid of OFMSW and protein rich poultry slaughterhouse waste.

For the pilot plant effluent from experimental period 2, upon the addition of "concentrated animal fat and protein" substrate, a significantly longer retention time until total digestion will be necessary. With both samples 2/1 and 2/2, a high methane productivity up to $0.33\,m^3.m^{-3}.d^{-1}$ could be achieved. After 30 days of additional digestion the methane productivity clearly dropped. Due to these facts a second phase digester should be implemented for a retention time of approximately 30 days. Within 30 days the methane yield can be increased significantly by 8.84 percentage points of total yield for sample 2/1 and 7.76 percentage points for sample 2/2. The additional COD degradation further stabilised the effluent. Data of additional methane yield and COD degradation are listed in Table 3.

Although the methane production for sample 2/2 decreased after 30 days, the methane generation continued longer than 78 days. This extended period of methane production in batch experiments could be due to a high content of accumulated harder degradable components or an inhibited anaerobic digestion process. NH_4-N, unionized ammonia and volatile fatty acids were detected in high concentrations, but not necessarily inhibiting

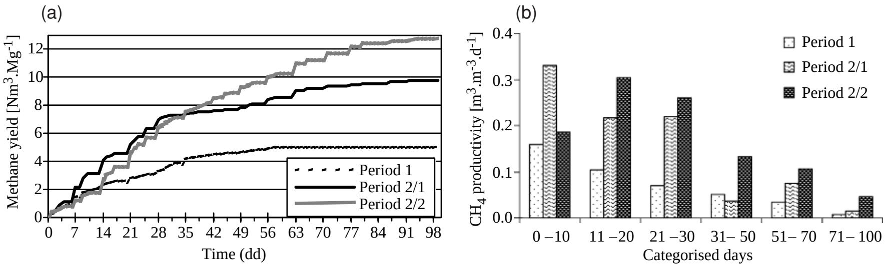

Figure 4 a) Cumulative methane yield of batch test experiments (period 1, 2/1 and 2/2) based on Mg pilot plant effluent b) Methane productivity for categorised retention times for the monitoring of the second digestion phase (periods 1, 2/1 and 2/2)

Table 3 Batch test experiments: methane yield, and additional methane production and COD reduction based on total digestion

	Methane yield ($Nm^3_{CH4}.Mg^{-1}_{FS}$)	Additional methane production (% of pot. yield)	Additional methane production within 25 days (% of potential yield)	Additional COD degradation (% of pot. yield)
Period 1	5.03	7.7	5.12	5.4
Period 2/1	9.76	11.7	8.84	8.9
Period 2/2	12.86	15.4	7.76	11.7

concentrations, during the continuous experiments in half technical scale. In experimental period 2 an accumulation of propionic and other volatile fatty acids with an odd-numbered chain was observed. Thus the monitoring of VFA decomposition and pH development during batch experiments was a further task in this study to assess the quality of the achievable end-product.

The effluent of experimental period 2 on day 48 (2/2) contained high acetic and propionic acid concentrations. During batch tests the acetic acid concentration was almost completely reduced in the same time period as it was observed in the final non-feed period of the continuous experiments (Figure 3a). Propionic acid accumulated during period 2 (up to 9,000 $mg.l^{-1}$). During 99 days of batch digestion the propionic acid was only reduced by 15%. It is well documented that organic acids having an odd numbered C chain may be degraded much more slowly than acetic or butyric acid with even numbered C chains (Shin *et al.*, 2003). Figure 5 shows that butyric acid increased during the first days and was almost totally reduced afterwards, whereas 30% of the initial concentration of the C-5 valeric acid was still remaining after 99 days.

Since this study should assess the feasibility of an entire anaerobic co-digestion process for the investigated substrates, different reasons for the low degradation rate of odd-numbered VFA have been evaluated.

Nonfavourable hydrogen partial pressures. The odd-numbered (C3) propionic acid is more difficult to degrade because its decomposition is only possible in a small thermodynamic range. As the propionic acid cannot be split into two acetic acid molecules, furthermore, this results in the longest digestion time for all VFAs. For this reaction sequence a hydrogen partial pressure of $<10^{-4}$Pa is mandatory (Cord-Ruwisch *et al.*, 1997; Schink, 1997). The increase of C3–C5 VFA is the response of the bacteria to increased hydrogen concentration in the liquid (Schink, 1997). In a technical-scale plant a re-inoculation may stimulate and accelerate the propionic acid degradation.

Unionized ammonia inhibition. During pilot plant experiments the unionized ammonia did not exceed 1,200 $mg.l^{-1}$ and this concentration effected no process inhibition.

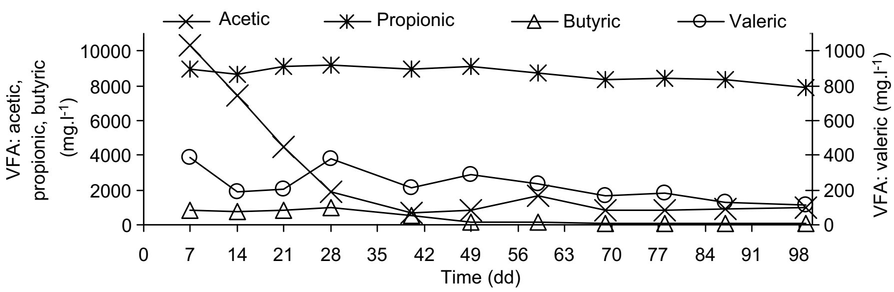

Figure 5 Development of volatile fatty acid concentrations during batch experiment – period 2/2

In batch test vessels few remaining protein components were broken up and ammonia released into solution. As the pH did not increase this slight increment of NH_4-N did not cause a severe increase of unionized ammonia and therefore nitrogen related process conditions did not change. There may have been a selective inhibition on propionic acid degrading bacteria populations (Angelidaki *et al.*, 1993; Schink, 1997).

Continuous supply of valeric and propionic acids from β-oxidation of long chain fatty acids. Valeric and propionic acids are the end-products of β-oxidation of odd-numbered long chain fatty acids. Due to sustained decomposition of long chain fatty acids, catalysed by acetyl co-enzyme A, valeric, butyric and propionic acids were continuously supplied (Bryant, 1979; Winter and Zeller, 1990; Schink, 1997). As previous studies showed, this can be demonstrated by an increase of butyric and valeric acids in the first weeks of the batch test (Shin *et al.*, 2003).

Direct inhibition of long chain fatty acids. The direct inhibitory effect that long chain fatty acids may possibly have was not investigated in further detail in this study. It is reported that in particular high concentrations of oleic and linoleic acids inhibit the methane production on acetate (Hanaki *et al.*, 1981). Accumulation of long chain fatty acids was proposed to be the main factor which affects the recovery of the process. Furthermore, similar studies suggest that the inhibition of propionate degradation by long chain fatty acids constituted the rate-limiting step in batch assays (Salminen *et al.*, 2000).

The risk of acidification caused by accumulation of VFAs and the rapid concomitant drop in pH (Kus and Wiesmann, 1995) is avoided because of the increased buffer capacity of NH_4-N. The expected increase of pH due to elevated ammonia concentrations was prevented because of the low pH of the substrate. Thus the unionized ammonia concentration did not exceed inhibitory levels. This treatment "equilibrium" works well as long as all intermediate products are decomposed completely and do not accumulate. An imbalance in the system will not cause a sudden failure as there will always be easily degradable components available for the microorganisms. Therefore, the high content of concentrated animal proteins and fats effected no instantaneous inhibition and as a result the measured methane yield was high and satisfactory.

The accumulation of less degradable and intermediate components consistently occurred but was accelerated under shock load conditions (data not shown). This imbalance may only be detected by regular analysis of process parameters such as VFA and COD (Björnsson, 2000). In full technical scale a constant high concentration of VFA might be no major problem but a sustained accumulation of intermediates can be avoided by adjusting the substrate mixture. Even if the process requires a final second digestion phase without additional substrate feed a fast decomposition of accumulated intermediates is not guaranteed. In particular C3–C5 carboxylic acids need special process conditions and time to be digested completely. This is not only documented in this study; comparable results were found during long-term monitoring of similar Austrian full-scale plants (data not shown). A biomass re-inoculation from a totally digested effluent may accelerate the decomposition in such a post digestion phase.

In an industrial-scale plant that is operated under similar conditions, all of the mentioned influences will be superimposed. It has to be considered if total degradation of propionic acid is essential. The achievable methane yield is not substantial, but incompletely degraded though still potentially available components may cause problems of downstream treatment options such as odour emissions in aerobic composting or in direct fertilizer use. Undegraded volatile organic components may also restrict ammonia recovery in a stripping process, due to contamination of the product by organic substances. This will negatively impact the end product quality.

Conclusions

Anaerobic digestion is a forward-looking technology for the treatment of most highly concentrated organic waste streams. Especially animal by-products from slaughterhouses and meat processing industry are well suited for anaerobic treatment. The European ABP regulation gives the legal basis for all treatment concepts. A number of studies identified the prospects and risks of ABP treatment (Salminen and Rintala, 2002). A risk that should not be underestimated for process stability is the high nitrogen and fat contents of ABP (Mata-Alvarez *et al.*, 2000; Salminen and Rintala, 2000). Mono-digestion in laboratory scale and in pilot plants, as well as first well documented experiences from full technical scale plants in Austria, showed that process instability cannot be excluded (data not shown).

In this study the feasibility of using two new substrates and their combination was tested for co-digestion. After characterising the waste streams in the laboratory the digestion process was monitored in half technical scale experiments to deliver recommendations for a full technical scale implementation. The co-digestion of easy available, partly hydrolysed organic wastes (pressed liquid) and protein rich waste fractions from slaughterhouses and the meat industry (blood and carcasses) is highly advisable.

The co-digestion of OFMSW and ABP in systems with a high organic load with a large variation in substrates will be common in future and may create an economically feasible opportunity for organic waste treatment. The gas yield of the waste streams can be increased, inhibition and process imbalances of mono digestion strategies can be avoided and a good quality end-product can be achieved if some key demands are met: sufficient, but technically feasible and economic HRT in phases 1 and 2 to break down harder degradable components of ABP wastes. Attention must be paid to the protein and fat content of the substrate to avoid possible inhibitions. A regular monitoring programme is mandatory for such high loaded systems. Finally, nitrogen recovery directly out of the process may increase the stability and productivity of the anaerobic digestion process even further.

Acknowledgements

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2. The influence of energy crop substrates on the mass-flow analysis and the residual methane potential at a rural anaerobic digestion plant

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The influence of energy crop substrates on the mass-flow analysis and the residual methane potential at a rural anaerobic digestion plant

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ABSTRACT

In a full scale anaerobic digestion plant exclusively operating on solid energy crops the mass-flows were analysed for two different substrate compositions over 583 d. The mono-fermentation of maize whole crop silage was compared to a mixture of maize and grass + clover silage. The two stage system required the input of dilution liquid guarantee digestion and agitation in the high loaded first stage (OLR: 5.50 $kg_{VS} \cdot m^{-3} \cdot d^{-1}$). Grass + clover demanded the double mass of process dilution liquid, which reduced SRT from 65 d to 34 d for each stage and leaded to an increased generation of Solid Digestion Residues by separation. Experiments showed that 70% of the Residual Methane Potential are caused by the 7% mass fraction of SDR. For maize and maize + grass + clover RMPs of 6.34% and 11.80% were observed, respectively. RMP can also be expressed as additional substrate input required for full granted operation. Thus, the mass stream analysis is used to determine mitigation strategies for RMP.

Key words | agricultural biogas plants, energy crops digestion, grass and clover, mass-flow analysis, organic loading rate, process dilution liquid, residual methane potential, solids separation

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INTRODUCTION

The anaerobic digestion technology is a multi-talent for renewable energy production. Various resources like wastewater, organic waste (OW), industrial and agricultural organic by-products and recently, cultivated energy crops are converted into bio-methane, which can be utilized in different energy services as process heat, electricity and transport fuel. These benefits can not only be realized in centralised units, but can also be organized in rural areas and new decentralised energy systems.

Granting systems for green-energy in many European countries led to a fast growing number of plants in particular in Austria and Germany. Recent evaluations of Austrian (Laaber 2005) and German (Weiland 2005) biogas plants show that an increasing number of plants is not longer coupled on manure digestion and that the application of mono-fermentation of energy crops is a fast increasing market. Earlier studies mainly focused on co-digestion with manure or OW (Zauner & Kuentzel 1986; Nordberg & Edström 2005; Lethomäki 2006; Nordberg *et al.* 2007; Svensson *et al.* 2007)

In this study the first Austrian biogas plant based exclusively on solid energy crops was monitored over a 2 years time period. Furthermore, this plant is located in a rural area in the eastern part of Austria, where the cattle farming was heavily hit by structural changes in the European agro-markets (Plieninger *et al.* 2006). Hence, fallow agricultural land increased, which is nowadays utilised for energy production. 11,000 t crop material from approximately 250 ha are converted into 4,300 $MWh \cdot a^{-1}$ electric energy and additionally 2,000 $MWh \cdot a^{-1}$ of thermal energy are used in the process or sold to the local district heating system. Maize is currently the most popular energy crop for anaerobic digestion, but its cultivation requires moderate temperature and water conditions and advanced agro-technology. Grass and clover are said to be alternative

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crops in the future, especially in alpine, rural and medium developed areas, like the one presented. Furthermore, the higher and different trace-element content of grass and clover (Nordberg *et al.* 2007) may compensate the supposed disadvantages of maize mono-digestion plant and may increase their performances (Weiland 2006).

At the monitored plant (Figure 1) the solid substrate fermentation technology is used to convert grass and clover together with maize into biogas. The fermentation is operating under thermophilic conditions of 49.5 °C and takes place in two 1,500 m^3 reinforced concrete tanks. The main fermenter (MF), is fed with the substrate fully automated 72 times per 24 h. The secondary fermentation step (SF) is connected in series, to establish optimised cascade degradation. Both tanks are equipped with horizontal paddle stirrers. Due to the high VS concentration of the utilized energy crops, a certain dilution is required to guarantee sufficient mixing and the best contact condition between substrate and micro-organisms. The process liquid for dilution is composed of the liquid fraction of separated digestion residue, surface and bunker silo runoff and the intense rainfall run-off. No input of manure or other liquid substrate is provided.

The most important issue for research and development at this new anaerobic digestion system is the management of the recirculation liquid to meet the following parameters: (I) full supply of contracted energy; (II) controlled microbiological process and stabilisation of total solids (TS) concentration in both fermenters; (III) high CH_4 yield and efficiency of substrate energy conversion. For a technical and economical success all three issues are extremely important. The limited market price for energy and the increasing prices for energy crops, push high pressure on the achievement of high efficient anaerobic digestion plants.

In this study the mass flows and energy balance from the full size plant associated with in-depth laboratory experiments over a 583 day period are presented. The focus of interest is set on two substrate input cases. In case 1 the system was operated with 100% maize whole crop silage. Case 2 compares the system parameters when 52% grass and clover silage and 48% maize were fed in. The evaluation clearly shows differences in required process liquid, productivity in the fermentation cascade and methane yield from volatile solids (VS).

MATERIAL AND METHODS

Mass flow analysis

Mass flow analysis are a tool to describe and analyse complex systems. They allow to print and model all mass and energy flows as well as single processes and figure out the relevant ones for a certain research question. Goal-oriented actions and optimisations can be derived consequently (OENORM S 2096–1, 2005). All streams and processes are identified first and then delimited spatially and temporally to describe alternating coherences.

Full scale system data collection and processing

Mass flow data of solid, liquid, and gaseous streams are measured on site at the full scale plant and stored in a data

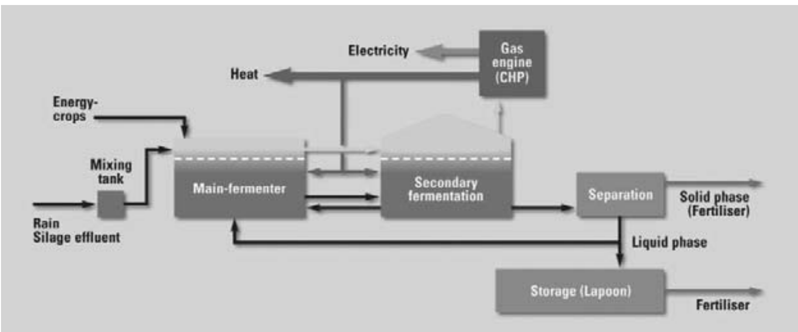

Figure 1 | Process scheme of the studied biogas plant. The white circles indicate the measuring points for residual methane potential evaluations.

archive on daily basis. All installed equipment is industrial standard and, if required, calibrated regularly.

Sampling

Fermenter 1 and 2 were sampled weekly taking 20 l from a sampling valve into a bucket, from which a quota of 0.5 l was taken out and cooled immediately down to 4 °C. Solid (SDR) and liquid digestion residues (LDR) were sampled every 42 days according to similar procedures.

Laboratory experiments

Volatile fatty acids (VFA) were analysed using High Performance Liquid Chromatography (Agilent 1100, RI-Detector). Total nitrogen (TKN), total ammonia nitrogen (TAN), pH, total solids (TS) and volatile solids (VS) were determined using *Standard Methods* (APHA 1998). VFA, TAN and TKN have been used for process control, but are not displayed and discussed in this study.

Residual methane potential

The methane potential of all used substrates and the residual methane potential of the fermenter 1 and 2 contents, the LDR and SDR were evaluated through anaerobic batch degradation tests every 70 days. These tests were carried out according to the modified standards DEV S6, DIN 38 414-S6 in 500 ml bottles at 35 °C. Samples from LDR could be used directly, samples from fermenter 1 and 2 have to be diluted 1:2 with tap water for adequate stirring. Substrates and SDR were inoculated with adapted bacterial biomass. All batch fermentation tests were carried out in three replicates and continued until no more gas production was observed (up to 110 days).

Calculation of performance parameters

Mass flow data (wet weight, WW) from the full scale plant, the regularly measured TS and VS concentrations and results from batch fermentation tests allowed to develop and derive the complete mass (WW, TS, VS) and energy (CH_4) flow system on a daily basis. The energy conversion performance was measured on the balance of total VS input and output as well as the VS degradation rate (DR). These are two different parameters: For the $VS_{IN} - VS_{OUT}$ balance the mass of VS_{IN} is 100%, whereas for the DR the technically maximum achievable methane yield of a certain substrate in a batch degradation tests is fixed as the 100% baseline. For the second model the methane produced in the plant the methane yield from batch test of SDR and LDR must equal the methane yield of the substrate out of batch degradation tests in a daily mass flow balance. This fact has been used as a control parameter for the mass flow system. The difference between these two degradation parameters is the part of VS that was not digestible biologically, but is potentially utilizable energy.

RESULTS AND DISCUSSION

Substrate input and case classification

For this study the mass flow data base was enquired for 2 different substrate input cases. Figure 2 shows the substrate input and methane yield for 583 days as weekly average. Case 1, which is marked white on the x-axis, refers to the time periods when nearly 100% maize was used as substrate (5 time blocs; 272 days). Case 2 (black) represents a substrate mixture of 48% maize and 52% grass + clover (5 time blocs; 311 d). All performance data have been averaged statistically for

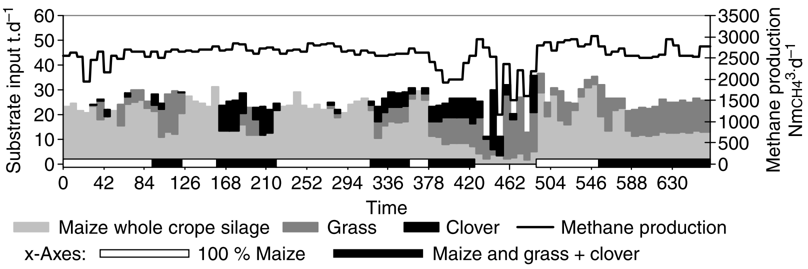

Figure 2 | Substrate input as weekly average in t·d^{-1} and methane production in Nm^3·d^{-1}. The time periods are marked on the x-axis: White for case 1 (100% Maize) and black for case 2 (Maize and grass + clover).

Table 1 | Mass streams of input, recirculation, effluent and biogas output in $t{\cdot}d^{-1}$ for both examined cases

	Maize				**Maize + Grass, Clover**			
	$t{\cdot}d^{-1}$		$t_{VS}{\cdot}d^{-1}$		$t{\cdot}d^{-1}$		$t_{VS}{\cdot}d^{-1}$	
Input								
Substrate								
Maize Silage	26.02	(2.85)	7.93	(0.68)	12.41	(0.98)	3.74	(0.57)
Grass, Clover	~0.00		~0.00		13.44	(0.86)	4.41	(0.44)
Process Liquid	17.53	(2.31)	0.28	(0.21)	36.34	(3.85)	0.98	(0.11)
Summed Input MF	43.95		8.21		62.19		9.13	
Separation and Output								
Solid digestion residues	2.31	(0.63)	0.49	(0.08)	3.80	(5.30)	0.78	(0.06)
Liquid digestion residues	30.21	(2.78)	1.06	(0.08)	47.29	(1.66)	1.65	(0.12)
Output into Final storage lagoor	27.76	(2.30)	0.55	(0.10)	29.19	(2.55)	0.76	(0.21)

each bloc first and then for the whole case. Figure 2 also displays that the methane production was constant and not affected by changing substrate mixtures. Due to maintenance and system tests the data from day 527–580 was excluded from the calculations.

Mass flow analysis

The mass streams have been calculated on a daily basis and then averaged for Cases 1 and 2. Table 1 displays the most important streams in $t{\cdot}d^{-1}$ and $t_{VS}{\cdot}d^{-1}$ including the standard deviation in brackets. In Figures 3 and 4 all mass flows are shown as SANKEY diagram, in which the amplitudes of the streams represent the masses. In both cases the system has been operated as scheduled in cascade degradation, separation of solid and liquid digestion residues and of parts of LDR as dilution liquid.

After separation of SF output the SDR counts for 7.1% of the separated WW in case 1 and 7.4% in case 2, but for 31.62% and 32.09% of the VS separated. The SDR is stored under aerobic conditions and finally used as high quality fertilizer in horticulture. The LDR is either pumped directly into the final

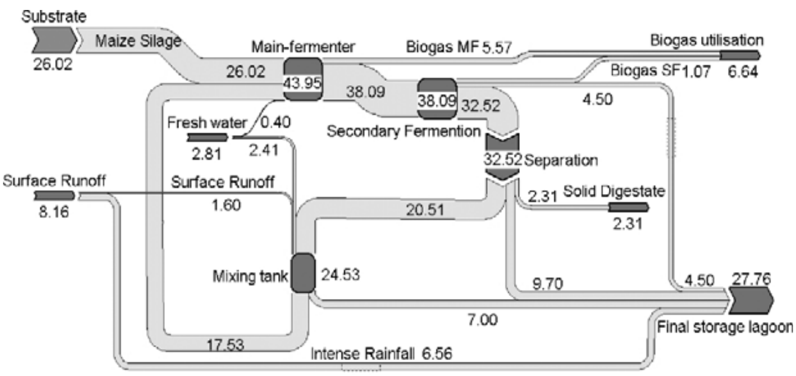

Figure 3 | Complete mass flows averaged for case 1 (100% maize silage substrate) in $t_{WW}{\cdot}d^{-1}$.

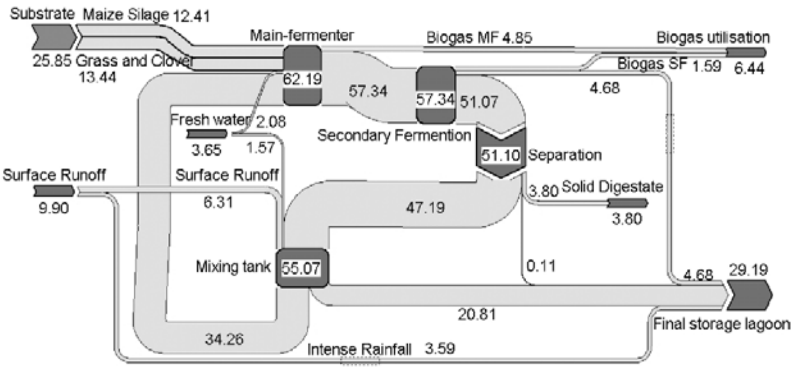

Figure 4 | Complete mass flows averaged for case 2 (Maize silage and grass + clover substrate) in $t_{WW} \cdot d^{-1}$.

storage lagoon (FSL) or into a mixing tank (MT). There, it is mixed with surface runoff from the solid covered areas. Parts of the process liquid are pumped into the MF or disposed into FSL. In terms of separated VS 27.78% sustained in the process and 38.19% are sorted out into the lagoon in case 1. When grass + clover are utilised, (case 2) 38.65% of VS are re-fed into MF and 30.27% are disposed.

Dilution liquid management

The dilution liquid mass stream is the key stream for the operation of this system. It should on the one side be as small as possible because it lowers the SRT for the substrate in the whole cascade. On the other side the discharge is determined by the TS concentration in MF, that has not have to excess 10%. This limit was tested out the maximum TS for adequate agitation for the installed system and is also described in other case studies (Weiland 2006; Nordberg & Edström 2005).

Figures 3 and 4 show, that the dilution stream had to be doubled from 17.53 $t \cdot d^{-1}$ to 34.26 $t \cdot d^{-1}$ when grass + clover were used additionally to maize silage. The reason for increasing the process liquid is that fibrous substrate, such as grass and clover, has a large impact on the rheological properties of the digestion liquid. The VS of grass and clover consists of higher contents of celluloses, hemicelluloses and lignin instead of starch and soluble sugars (analytical data not shown) (Lethomäki *et al.* 2006; Martinez-Perez *et al.* 2007).

The complex organic structure decelerates the degradation kinetics of the VS, which can be clearly demonstrated in batch degradation tests (Figure 5). Although the cumulative methane yield was similar (440 $Nm^3_{CH_4} \cdot t^{-1}_{VS}$) for maize silage and wet grass + clover silage, 50% of maximum was yielded at day 7 for maize and at day 12 for grass and clover silage. If the grass or clover was harvested too late, the degradation kinetics is decelerated further and total yield was reduced by 20% and the 50% yield was measured on day 16. Model parameters for the hydrolysis and methanogenesis kinetics are under way for those complex crop substrates and will be implemented in ADM1 (Batstone *et al.* 2002), to improve mathematical modelling of such data sets.

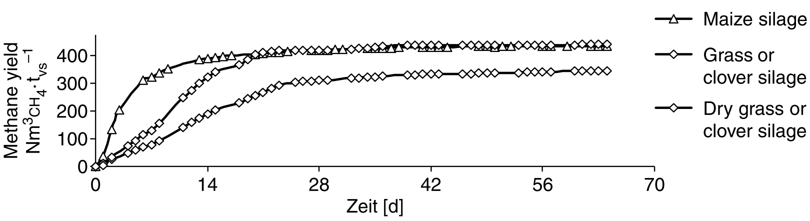

Figure 5 | Batch degradation of Substrates utilized in full scale plant showing different degradation kinetics.

Reduced hydraulic retention time and high Organic Loading Rate (OLR)

The additional input of recirculation liquid in MF leads to a reduction of calculated HRT. Per definition the HRT equals the solid retention time (SRT) in a completely stirred tank reactor (CSTR). Thus, the SRT for the energy crop substrate input is reduced equally. In case 1 the SRT for the maize silage had been derived 63.40 d in MF and 65.08 d in SF (128.48 d in total) (Table 2). In case 2 the SRT is reduced to 33.73 d in MF and 34.76 d in SF. Compared to data of recent studies the median SRT for modern agricultural biogas plant in Austria is 139 d (Laaber 2005) and for 2 stage systems the SRT is generally longer than 90 days in Germany (Weiland 2005).

The OLR of a biological system is defined as the mass of substrate input per m^3 of fermentation volume (FV). For a recirculation system it has to be stated whether the additional load of digested liquid is included in the OLR or not. Table 2 shows the calculation for both options and viewed substrate cases. The OLR of the MF was 5.19 and 5.35 $kg_{VS}\cdot m_{FV}^{-3}\cdot d^{-1}$ for Case 1 and 2 based on substrate input. This OLR is 57.8% higher than the Median of Austrian biogas plants. The high organic loading rate in all substrate input cases did not influence the microbiological process tability. VFA did not increase 250 $mg\cdot l^{-1}$ in MF and 200 $mg\cdot l^{-1}$ in SF over the whole review period. Nordberg & Edstörm (2007) achieved an OLR of 3 $kg_{VS}\cdot m_{FV}^{-3}\cdot d^{-1}$ in a recirculation system digesting energy crops. Svensson *et al.* (2007) achieved a VS reduction of crop residues of 80% at an OLR of 2.05 $kg_{VS}\cdot m_{FV}^{-3}\cdot d^{-1}$. Nordberg & Edström (2005) described an OLR of 5.5 $kg_{VS}\cdot m_{FV}^{-3}\cdot d^{-1}$, at a substrate mixture of 50:50 ley crop silage and municipal solid waste, but observed unstable conditions due to increasing VFA. Lethomäki (2006) investigated an OLR of 4 $kg_{VS}\cdot m_{FV}^{-3}\cdot d^{-1}$ at crop: cow manure ratio of 40:60 and a SRT of 16 d at a VS degradation of 40%.

Residual methane potential

For the evaluation of technical or energetic efficiency the rate of conversion of substrate energy into valuable energy services has to be optimised. Within the balance system border of the plant-site the degradation rate of VS-added has to be optimised and therefore the VS output and the residual methane potential have to be minimized. For Case 1 (100% Maize) a

Table 2 | The Organic Loading Rate (OLR), Hydraulic Retention Time (HRT) and Biogas Productivity (P) are calculated from full scale data base for case 1 and 2

	Units	Maize	Maize + Grass, Clover
Organic Loading Rate			
OLR MF: based on substrate input	$kg_{VS}\cdot m_{FV}^{-3}\cdot d^{-1}$	5.19 (0.45)	5.35 (0.29)
OLR MF: based on mass stream	$kg_{VS}\cdot m_{FV}^{-3}\cdot d^{-1}$	5.79 (0.45)	6.07 (0.51)
OLR SF: based on mass stream	$kg_{VS}\cdot m_{FV}^{-3}\cdot d^{-1}$	1.69 (0.49)	2.53 (0.54)
OLR: based on substrate input	$kg_{VS}\cdot m_{FV}^{-3}\cdot d^{-1}$	2.65 (0.25)	2.72 (0.28)
Hydraulic Retention Time			
SRT Main fermenter	d	63.40 (5.13)	33.73 (4.51)
SRT Secondary fermentation	d	65.08 (5.25)	34.76 (4.68)
Methane Concentration Biogas Productivity	Vol%	51.26 (0.49)	52.88 (0.53)
Main fermenter	$Nm_{CH_4}^3\cdot m_{FV}^{-3}\cdot d^{-1}$	2.96	2.43
Secondary fermentation	$Nm_{CH_4}^3\cdot m_{FV}^{-3}\cdot d^{-1}$	0.55	0.81

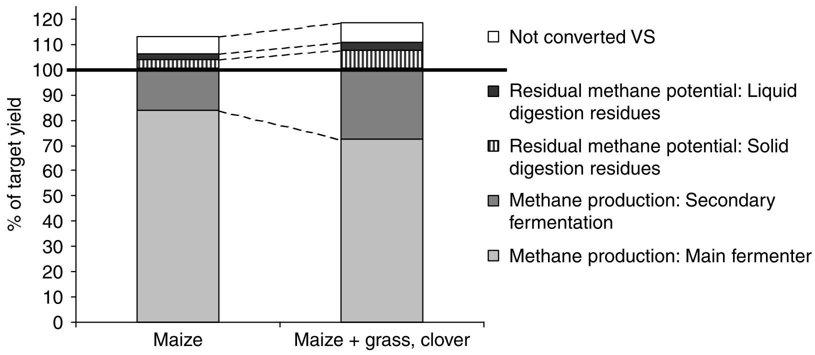

Figure 6 | Methane production in MF and SF for both cases to meet the 100% target line for full contracted operation. The segments above 100% can be expressed as the additional substrate input, which is required due to incomplete degradation of VS.

methane yield of 361.68 $Nm^3_{CH_4} \cdot t_{VS}^{-1}$ in the full scale plant was achieved. The CH_4 yield was reduced to 332.32 $Nm^3_{CH_4} \cdot t_{VS}^{-1}$ for Case 2 mainly due to shortened SRT and secondly due to lower methane potential of some grass and clover batches.

In the mass balance based on daily measurements, the slower degradation kinetics of grass and clover can be displayed on a shift of methane production towards the SF (Figures 4 and 6). In case 1, 84.18% of methane production takes place in MF. In case 2, only 72.38% CH_4 are converted within MF resulting in a higher portion of VS, which is discharged into SF for cascade degradation.

Consequently, a higher CH_4 potential in a complete stirred reactor (CSTR) leads towards a higher residual methane potential (RMP) in the SF effluent. The RMP was tested in batch degradation tests 10 times within the observation period for all effluents (MF, SF, LDR, SDR (Figure 1)). The RMP is tested under optimum conditions (anaerobic, 35 °C, 110 d) and can therefore not be directly related to potential green-

Table 3 | Methane production in Main Fermenter (MF) and Secondary Fermentation (SF) and residual methane potential (RMP) of solid (SDR) and liquid (LDR) digestion residues for both cases in $Nm^3_{CH_4} \cdot d^{-1}$ for the full scale plant

	Maize		**Maize + Grass, Clover**	
	$Nm^3_{CH_4} \cdot d^{-1}$	$Nm^3_{CH_4} \cdot t_{VS}^{-1} \cdot d^{-1}$	$Nm^3_{CH_4} \cdot d^{-1}$	$Nm^3_{CH_4} \cdot t_{VS}^{-1} \cdot d^{-1}$
Methane Production				
Methane Main fermenter	2272.00 (145.18)		1935.00 (291.96)	
Methane Secondary fermentation	427.00 (88.56)		640.00 (141.45)	
Methane Yield		361.68 (35.21)		332.32 (36.67)
Residual Methane potential				
Effluent Secondary fermentation	188.95 (21.30)		347.63 (31.89)	
Solid digestion residues	107.26 (74.41)	180.45 (19.34)	196.26 (92.92)	215.52 (20.79)
Liquid digestion residues	78.90 (4.32)	68.32 (5.78)	131.89 (11.25)	78.68 (6.02)
Input Final storage lagoon	50.12 (5.89)		74.78 (37.75)	

house gas (GHG) emissions of open stored digestion residues. GHG emissions are sensitive data that have to be measured accurately in specific designed test schemes.

Although, according to Table 1, the mass flow of SDR contributes only 10.90% of the disposed mass (SDR + LDR into FSL) in case 1, the RMP of SDR counts for 68.29% of total system effluent RMP. When grass and clover were used, the mass of separated SDR increased and represented 15.80% of mass flow and 72.42% of total RMP. Table 3 displays, that the methane production within MF and SF was nearly equal in case 1 and 2 and made a plant availability of 96.11% and 94.74% possible. In terms of t_{ww} disposed the RMP arises 7.42 $Nm^3_{CH_4} \cdot t^{-1}_{ww}$ and 8.96 $Nm^3_{CH_4} \cdot t^{-1}_{ww}$ for case 1 and 2, respectively.

Additional substrate input

An existing energy crop based biogas plant selling the energy for a fixed grant into the electricity grid cannot get more efficient by increasing the OLR or the biogas productivity as it is common for waste treating plant which have benefits from gate fees. Energy crop plants can only get more efficient in delivering 100% of contracted energy using as little purchased crop substrate as possible. Optimum substrate energy conversion and minimizing RMP are the key parameters. Figure 6 displays this optimisation calculation from the operators point of view. The y-axis marks % of targeted yield, which means that 100% represents the methane yield required for 100% CHP operation. To score 100% plant operation load a RMP has to be taken into account.

For 100% maize input a RMP of 6.34% of CH_4 production was assessed. This can either be expressed as 94% substrate degradation rate, or, from the operators view, 6.34% extra substrate input because of incomplete degradation. As remarked, 68.29% of RMP are generated by the SDR. Due to the doubled stream of dilution liquid in the case of grass + clover substrate the RMP increased to 11.80%, of which 72.42% are caused by SDR. In his study Weiland (2005) calculated the RMP in a broad range of 5–15% of biogas production for German biogas plants, which are all operated under lower OLR than the presented one. Lethomäki (2006) reports a RMP of 3–5% for cow manure based experiments. In the mass balance the degradation efficiency can also be stated as $t_{VS\,Input} - t_{VS\,Output}$. This view includes the "non convertible" VS and reduces the calculated VS degradation rate. In Figure 6 this portion of VS is represented by the top segment of the columns and counts for 11.11% and 13.90% of additional required substrate input in case 1 and 2, respectively.

CONCLUSIONS

A strategy for mobilising currently unused methane potentials has to have two targets. First of all the full scale continuous energy conversion has to be as close as possible towards the technically maximum. Secondly the potential of non convertible VS has to be accessed by adequate mechanical, thermal, chemical or biotechnological pretreatment techniques (Perez Lopez *et al.* 2005). Primarily, the process management on site has to be improved: in the presented case study the input of dilution liquid has to be minimized and consequently the discharge of incompletely degraded SDR by separation is reduced. One possibility for dilution in MF might be the use of fresh water instead of LDR. Beneficially, the SRT will be reduced less than currently because of the higher dilution factor of pure water. The predicted decrease of RMP has to be calculated against the costs of discharging the additional water as "fertiliser", which are derived from 3–5 €.t^{-1}. Figure 3 and Figure 4 show another dilution alternative: the surface runoff from precipitation on solid plant areas counts for considerable amounts of water that have to be disposed on site together with LDR. Surface runoff has to be included as good as possible within the recirculation scheme, because it causes only small reduction of SRT, but has a large beneficial effect on RMP. Although, the surface runoff is hard to manage as intense rainfall can hardly be forecasted, the management of surface runoff and better integration as recirculation liquid has been improved continuously to minimize the demand of LDR for recirculation.

The results of this study demonstrate that the exclusive digestion of solid energy crops is technically feasible, when the recirculation of process liquid (LDR) is managed properly. A change in substrate composition containing higher proportions of fibrous and celluloses substrates and celluloses leads to an increase in recirculation demand, which lowers the HRT and generates more solid digestion residues. Since the largest source of residual methane potential can be found in

SDR, the minimisation has to be the main goal. The RMP can also be expressed as non converted VS or as additional substrate input that has to be provided by the operator to meet the targeted energy output. Rising prices for energy crop substrates generate a high demand for process operation schemes to minimize the RMP.

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3. Enhancement options for the utilization of nitrogen rich animal by-products in anaerobic digestion

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Enhancement options for the utilization of nitrogen rich animal by-products in anaerobic digestion

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Abstract

This study focuses on the enhancement of an Austrian anaerobic digestion plant at a slaughterhouse site which uses exclusively animal by-products as substrate. High ammonia concentrations resulting from protein degradation cause a severe inhibition of the anaerobic microorganisms. For improving the current situation the COD:TKN ratio is widened by a) ammonia stripping directly out of the process and b) addition of a C source to the substrate. Different organic loading rates and hydraulic retention times were tested in continuous experiments to simulate new operating conditions. The results show that the addition of biodegradable carbon cannot improve the fermentation capacity. The reduction of ammonia boosts the digestion process: After reduction of TKN from 7.5 to 4.0 g kg^{-1} the initially high VFA concentration decreased and the COD degradation was improved by 55.5 %. Hence, the implementation of the new N reduction process facilitates either the increase of the OLR by 61 % or the reduction of the HRT by 25 %.

Keywords: anaerobic digestion, slaughterhouse wastes, ammonia recovery, COD:TKN ratio, inhibition,

1. Introduction

Animal by-products not intended for human consumption or animal feed according to the ABP-regulation (Reg. (EC) No. 1069/2009 – replacing the ABP directive 1774/2002) have a high potential in the production of methane in anaerobic digesters. Since the regulation is defining new treatment possibilities alternative pathways for the utilisation of slaughterhouse wastes were opened. Methane can be recovered and used in energy production to replace fossil fuel and process energy and CO_2-emissions are reduced thereby. In contrast to aerobic waste and wastewater processes most of the nutrients remain in the treated material and the end-product can be used as agricultural fertilizer. The high nitrogen content in the proteins of ABPs is challenging mono digestion processes.

Concerning industrial scale anaerobic digestion there is only fragmented information available about the limits of the utilization, the behaviour of high protein containing wastes, new process options and the integration of slaughterhouses and anaerobic digestion plants. (Edström et al., 2003; Lopez et al., 2006; Resch et al., 2006). Batch experiments and pilot scale applications of anaerobic digestion of slaughterhouse wastes are presented in literature. (Buendía et al., 2009; Hejnfeldt and Angelidaki, 2009; Siegrist et al., 2005; Wu et al., 2009; Shanmugam and Horan, 2009; Wang and Banks, 2003)

1.1 Full scale plant

This work investigates process enhancement options for a biogas plant running at a large Austrian slaughterhouse, slaughtering approx. 10,000 pigs a week. Colon, colon content, blood, fat, scrubber content from a dissolved air flotation, stomach content, process water and some ABP fractions from a close-by cattle slaughterhouse are used as substrates. Substrate characteristics of ABPs are outlined by Kirchmayr et al. (2007) and Tritt and Suchardt (1992).

Corresponding to the initial design of the biogas plant only parts of the waste stream from the abattoir can be used as substrate. The protein nitrogen of the substrate and further process constrains are currently limiting the fermentation process and the mass of ABP that is converted into renewable energy. Due to the high nitrogen content 50 % of the annual blood volume has to be discharged to a rendering plant. Outlook calculations show that the power production may be doubled if a process will be set in place which enables to utilize all available ABPs.

Kirchmayr et al., (2009) and Kirchmayr et al., (2010) described the full scale case in detail. Over 4 years monitoring period the plant has been operated at an organic loading rate (OLR) 1.07 kg m^{-3} d^{-1} COD (5.07 kg m^{-3} d^{-1} VS) in fermenter 1. Depending on the substrate composition ammonia-nitrogen (TAN) levels vary between 5.3 and 6.2 g l^{-1} in fermenter 1 and between 6.0 and 7.4 g l^{-1} in fermenter 2. This corresponds to a Total Kjeldahl Nitrogen (TKN) concentration between 7.1 and 8.5 g kg^{-1}. High levels of ammonia lead to high concentrations of volatile fatty acids (VFA,) indicate an imbalanced microbiological activity and facilitate foaming problems. Median concentrations of 5.92 g l^{-1} acetic acid (AA) (Max 11.82) and 5.30 g l^{-1} propionic acid (PA) (Max 6.67) have been measured over 4 years. The average COD:TKN ratio was calculated 8.49:1.

1.2 Ammonia Inhibition

Ammonia is produced from the microbial degradation of nitrogen containing compounds which are primarily proteins. Its function as an essential nutrient for the microorganisms (Gallert et al., 1998), as cause of complex process inhibition (Chen et al., 2008), the inhibition relation to the concentration of undissociated ammonia nitrogen (UAN) (Braun et al., 1981; Koster, 1986; Hansen et al., 1998; Sprott et al., 1985; Strik, 2006) are widely reported in literature. As a component of the buffer system it initially appears to stabilize the pH value by maintaining a high level of bicarbonate in the wastewater (Salminen and Rintala, 2002; Speece, 1996)

1.3 Ammonia inhibition and VFA degradation

Anaerobic digestion at higher ammonia concentrations easily leads to an accumulation of VFA. This compensates the alkaline effect of ammonia resulting finally in a decline of the pH particularly if the buffer capacity of the fermenter sludge is insufficient. In addition the sinking pH causes a bigger proportion of undissociated fatty acids that have a toxic effect on the microorganisms too (Angelidaki and Ahring, 1993; Hansen et al., 1998; Hansen et al., 1999; Koster and Lettinga, 1984; Wiegant and Zeeman, 1986). An exact ammonia concentration limit, which should not be exceeded, is difficult to fix as the toxic effect depends strongly on the respective fermentation conditions (pH, temperature) and the present bacterial strains. It has been reported, that methanogens are capable of adaption to high ammonia concentrations when increasing the concentration slowly over a longer period (Angelidaki and Ahring, 1993; Angenent et al., 2002). However, the methane production in such cultures is lower. On the other side, the tolerance to TAN and pH variations seems to be increased (Sung and Liu, 2003).

1.4 COD:TKN Ratio

A lack of nitrogen causes an insufficient utilization of carbon sources. On the other hand, too narrow C:N ratios are leading to an accumulation of ammonia in the fermenter sludge which may subsequently lead to an inhibition of the microorganisms. In order to reach an optimal biogas yield in the balance of sufficient nutrient supply and ammonia inhibition, a C:N ratio ranging from 16:1 to 25:1 is reported (Hills, 1979; Shanmugam and Horan, 2009). For overcoming ammonia inhibition Kayhanian (1999) recommended the adjustment of COD:TKN ratio or the dilution of the digester whereas Nielsen and Angelidaki (2008) suggested re-inoculation with digester liquid to increase the biomass concentration.

1.5 Experimental setup, goals and measured parameters

A new process design, to improve the process performance and to be prepared for a site enlargement, is proposed. In order to assess on the one hand the amplitude of the inhibition effect of ammonia on anaerobic digestion and on the other hand to test strategies for overcoming high ammonia concentrations, the digestion of a nitrogen rich substrate was tested under various COD:TKN ratios. Two different strategies were compared to the reference conditions in long time continuous laboratory investigation. Fig. 1 shows the current full scale plant, the considered implementation of a new process step and the derived laboratory test setup.

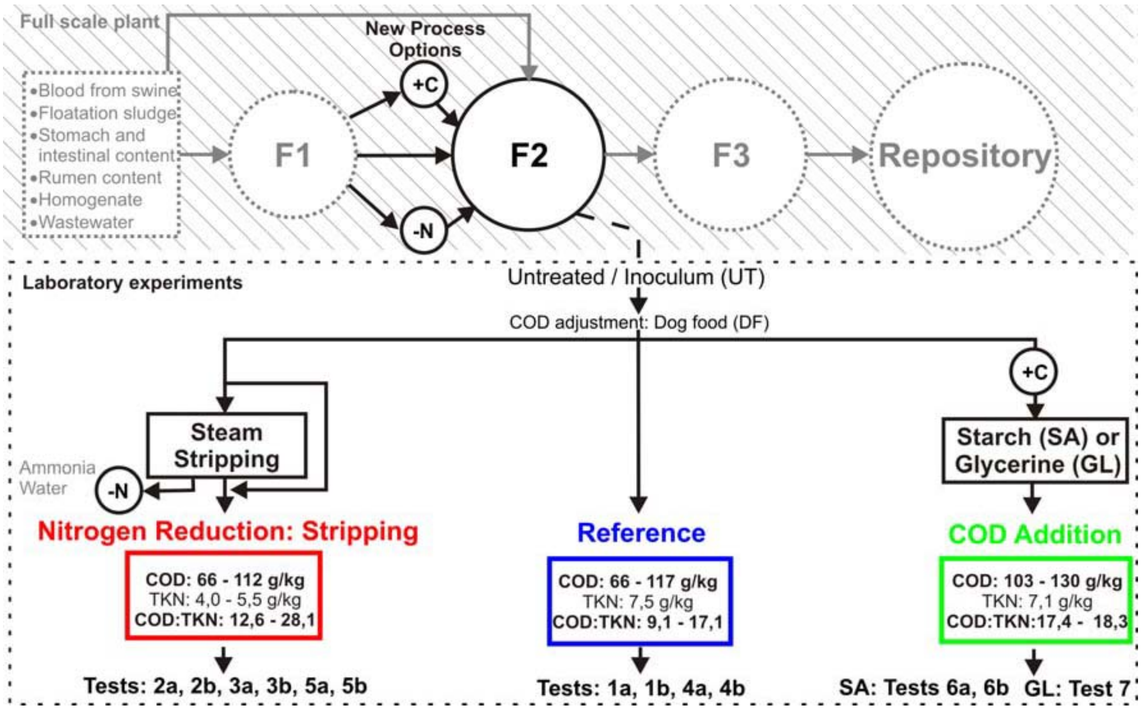

Fig 1.: Design of full scale plant and investigated enhancement options Nitrogen Reduction by Stripping, Reference and COD Addition in continuous laboratory experiments

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The first approach is to reduce the TKN in form of ammonia directly out of the process. A stripping unit might be placed after the first fermenter. In this study reduction and recovery of ammonia nitrogen was achieved by steam stripping of the digester content.

The second approach may be to increase the COD to widen the COD:TKN ratio using a carbon rich substrate like starch from energy crops or glycerine (industrially available by-product from biodiesel production). The results of continuous degradation tests should provide clear evidence whether the substrate treatment strategies have a positive effect on anaerobic digestion or not.

In order to provide comprehensive decision criteria for the setup of a new process the experimental design has to cover a broad range of process parameters. Therefore, the parameters TKN, Organic Loading Rate (OLR), Hydraulic Retention Time (HRT), COD:TKN were chosen to define the set of continuous degradation tests.

2. Material and Methods

2.1 Inoculum and Substrate

The inoculum and the substrate for the laboratory tests were taken out of fermenter 2 of the full scale biogas plant. The parameters are shown in Table 1. To balance fluctuations and to compensate pre-digestion the substrate was supplemented with COD from dog food, which has similarly characteristics as ABPs. It was also treated by steam stripping in order to reduce the ammonia content without changing the COD. The carbon sources used for several test setups were added to the basic substrate.

Table 1
Chemical Parameters of Inoculum and Substrate from fermenter 2 of full scale plant

pH		8,08
TKN	g kg^{-1}	7,69
TAN	g l^{-1}	6,61
COD	g kg^{-1}	51,89
Acetic acid (AC)	g l^{-1}	5,66
Propionic acid (AC)	g l^{-1}	5,53
Remaing acids (RA)	g l^{-1}	1,54

2.2 Steam stripping

A laboratory scale, continuous steam stripping experimental plant was designed. In the literature steam stripping of ammonia is reported as being more effective than using air (Sattler, 2001; Hwang et al., 1992). Changes in substrate composition due to the stripping step were measured and documented.

2.3 Test setup and process control

Batch degradation tests. The degradation performance of the chosen attempts was assessed in batch degradation tests in advance. These tests were carried out according the guideline VDI 4630.

Continuous stirred tank reactor experiments. The tests were carried out in stirred 2,000 ml bottles at 39 C. The produced biogas was washed in Sodium Hydroxid (NaOH) to remove CO_2. All tests were conducted in duplicates and the length of experiments was at least three times the HRT. The tests were fed on daily basis.

*Laborator*y Analysis. Volatile fatty acids (VFA) were analysed using High Performance Liquid Chromatography (Agilent 1100, RI-Detector). Total Nitrogen (TKN), Total Ammonia Nitrogen (TAN), pH, and Chemical Oxygen Demand (COD) were determined using standard methods (APHA, 1998).

Process Control. The yield of CH_4 was measured continuously using automatic counter system. pH was measured every second day, VFA was analysed weekly and COD, TKN and TAN on 14 days basis.

2.4 Parameter of experimental design

The future enhancement considerations for the full scale plant define the setpoints for TKN, COD:TKN, OLR and HRT. Table 2 shows the parameters of the full set of 13 continuous degradation tests.

Total Nitrogen (TKN). The nitrogen content in the substrate of the Reference tests (1a, 1b, 4a, 4b) was set at 7.5 g kg^{-1}, which corresponded to the conditions in the full scale plant. For the Stripped tests the TKN was reduced to 5.5 and 4.0 g kg^{-1} (2a, 2b, 3a, 3b, 5a, 5b). These levels are in the range of industrial biogas plants in Austria and Germany and should improve the COD reduction and process stability. The TKN reduction was tested at two levels to support the design and development of the nitrogen recovery process.

COD:TKN ratio. Due to the removal of nitrogen or the addition of COD the COD:TKN ratio was widened. The close ratio of 9.1 to 9.8 in the Reference tests is assumed to be a major cause for process instabilities. By reducing N or adding C the ratio was widened to 12.3 - 28.1. As the tests attempts Reference and Stripping only differed in their nitrogen levels, the comparison of these two should reveal a possible toxic effect of ammonia. Stripping (TKN 4) and Glycerine/Starch were equal in their COD:TKN ratios, but differed in their nitrogen levels as Glycerine had the same TKN content as Reference. Hills (1979) reported an optimal methane yield at a C:N ratio of 25 by using different mixtures of dairy cow manure, glucose, and cellulose.

Organic Loading Rate (OLR). In industrial scale plants the OLR is the most used parameter to link between process stability and economic needs. The OLR at the focussed plant is limited by the nitrogen content of the substrate. The OLR was set between 1.8 and 3.85 kg_{COD} m^3 d^{-1} varying the both COD concentration of the substrate and the HRT. The addition of corn starch or glycerine to widened the COD:TKN ratio increased the OLR (6a, 6b, 7). The increase of substrate input in the full scale plant was simulated by enhancing the feed concentration using a mixture of starch and lard (4a, 4b, 5a, 5b).

Hydraulic Retention Time. As the size of the full scale plant is fixed, the capacity can be increased either by heightening the OLR or accelerating the microbiological activity. If this faster degradation can be achieved after the implementation of a nitrogen removal unit was tested by the reduction of HRT from 40 days to 30 days. All tests were carried out at both HRTs.

Table 2
Set points for continuous tests

		Parameters				
		TKN setpoint	HRT	OLR	COD / TKN ratio	COD Feed
		g kg^{-1}	d	kg_{COD} m^3 d^{-1}		g kg^{-1}
1a	Reference	7,5	40	1,80	9,7	73
1b			30	2,21	8,8	66
2a	Stripped	5,5	40	1,80	13,3	73
2b			30	2,21	12,6	66
3a	Stripped	4,0	40	1,80	18,3	73
3b			30	2,21	17,1	66
4a	Reference, high OLR	7,5	40	2,90	17,1	117
4b			30	3,85	17,1	115
5a	Stripped, high OLR	4,0	40	2,90	28,1	112
5b			30	3,71	28,1	112
6a	Added COD - Starch	6,1	40	2,90	18,3	112
6b			30	3,71	17,4	103
7	Added COD - Glycerin	7,1	40	2,90	17,5	124

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2.5 Performance Parameters

The objective for the enhancement of the full scale plant is a stable digestion process, a high COD degradation rate and consequently a high Methane yield. These parameters were used to evaluate the performance of the laboratory tests. The VFA concentration, pH and UAN were chosen to assess the stability of the digestion process. The UAN was calculated according to Lide (2009) and Kayhanian (1999). New data based on simulation models derived that this equilibrium calculation may overestimate the concentration of UAN in digester environments (Hafner et al., 2009; Kirchmayr et al., 2010).

3. Results and Discussion

3.1 Preliminary Batch Tests

Before starting the continuous tests the assumptions were tested in batch tests. The results are presented in Resch et al. (2007). The methane yield and COD reduction increased when the nitrogen content was reduced. The reduction of TKN to 4 g kg^{-1} led to the best degradation results of COD from 50.68 g kg^{-1} to a minimum 29.45 g l^{-1} after 76 days. Thus, the final value was defined as non degradable COD. Analysis of the remaining organic components showed that fibres from gut and rumen content are not fully degradable under anaerobic conditions. Inmaculada et al. (2009) also reported a high inert fraction in rumen content that reduced the COD removal in batch co-digestion experiments.

3.2 Continuous Tests

The feasibility of exact set-points of process parameters in the laboratory over a long period of time and the application of industrial ABPs gave a qualified combination of a controlled test environment in the laboratory and the simulation of full scale conditions. The results of all 13 attempts are presented in Table 3 and Fig. 2.

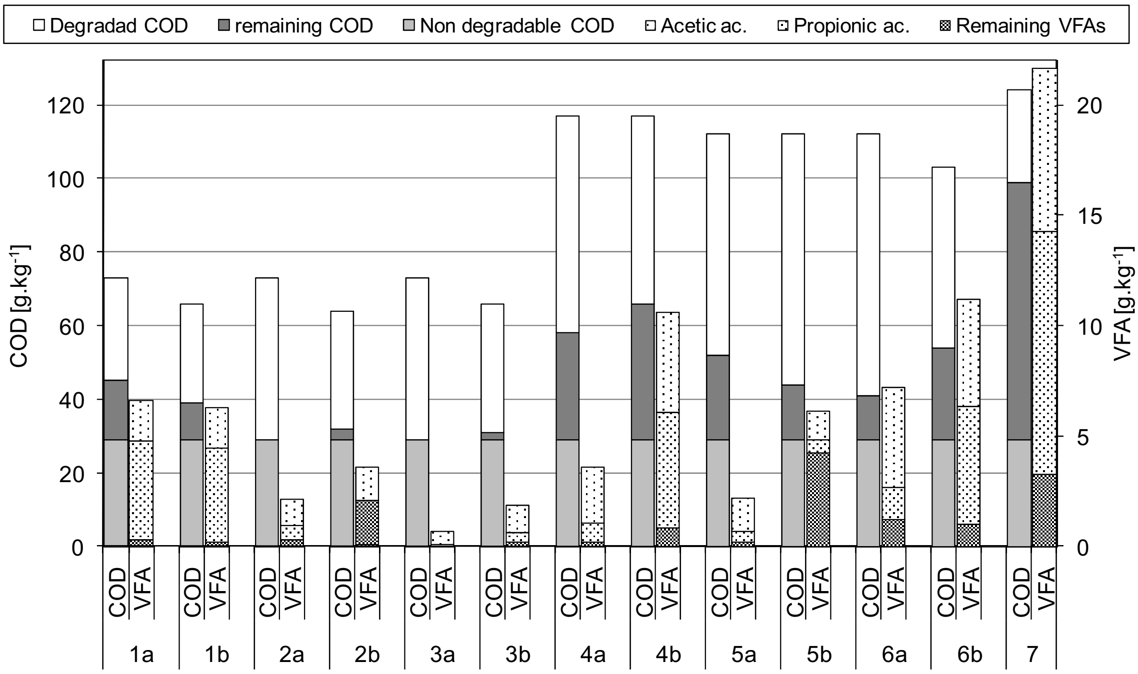

Fig 2.: COD degradation and volatile fatty acid concentration of 13 continuous degradation tests. Data represent median values of the last retention time. Remaining VFAs sum up butyric a., iso-butyric a., valeric a. and iso-valeric a.

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Table 3

Performance results of 13 continuous degradation tests. Data represent median values of the last retention time.

			COD				Y_{CH4}	Volatile Fatty Acids (VFA)				Nitrogen			
		TKN setpoint	COD In	COD Out	COD degradation	COD degr. of degradable COD	Methane Yield	Acetic Acid (AC)	Propionic Acid (PA)	Remaining Acids (RA)	pH	TAN	UAN	UAN min	UAN max
		$g \cdot kg^{-1}$	$g \cdot kg^{-1}$	$g \cdot kg^{-1}$	%	%	$Nm^3 \cdot t_{COD}^{-1}$	$g \cdot l^{-1}$	$g \cdot l^{-1}$	$g \cdot l^{-1}$		$g \cdot kg^{-1}$	$mg \cdot l^{-1}$	$mg \cdot l^{-1}$	$mg \cdot l^{-1}$
1a	**Reference**	**7,5**	**73**	**45**	**38,4**	**63,4**	**193**	**1,87**	**4,49**	**0,27**	**7,98**	**6,62**	**968**	**716**	**980**
1b			66	39	40,9	73,9	188	1,85	4,23	0,20	7,94	7,04	757	587	781
2a	Stripped	5,5	73	29	60,3	100,0	271	1,19	1,65	0,27	7,88	4,95	555	528	627
2b			66	32	51,5	91,4	193	1,48	1,98	0,10	7,87	4,62	542	449	549
3a	**Stripped**	**4,0**	**73**	**29**	**60,3**	**100,0**	**293**	**0,58**	**0,09**	**0,00**	**7,81**	**3,45**	**245**	**176**	**354**
3b			66	31	53,0	94,6	213	1,23	0,43	0,20	7,74	3,43	264	247	275
4a	Reference, high OLR	7,5	117	58	50,4	67,0	309	2,56	3,15	0,19	7,85	5,68	395	357	454
4b			115	66	42,6	58,0	223	4,50	5,21	0,86	7,76	6,67	506	472	609
5a	**Stripped, high OLR**	**4,0**	112	44	60,7	81,9	315	1,49	0,51	0,17	7,68	3,46	235	221	330
5b			**112**	**52**	**53,6**	**72,3**	**279**	**4,22**	**0,62**	**1,27**	**7,37**	**3,20**	**124**	**98**	**190**
6a	Added COD - Starch	6,1	112	41	63,4	85,5	299	4,54	1,45	1,20	7,68	5,06	318	274	491
6b			103	54	47,6	66,2	201	4,88	5,30	1,01	7,68	6,20	249	202	426
7	Added COD - Glycerin	7,1	124	99	20,2	26,3	41	7,38	11,03	3,25	7,55	6,49	556	342	733

3.2.1 Reference tests for the full scale plant

Different operating conditions for the full scale plant without optimization procedures were simulated in Reference test setups. The initial process parameters at an HRT of 40 days (1a) were well-known from the monitoring of the full scale plant over years. After a short start-up phase the COD content was reduced by 38.4 %. This corresponded to a 64.5 % degradation of the degradable COD and a methane yield Y of 193 Nm^3 t_{COD}^{-1}. Due to the high nitrogen content the pH was measured constantly at 7.98. The measured TAN concentration of 6.62 g l^{-1} was 88 % of TKN and resulted in a calculated UAN level of 968 mg l^{-1}. Although the tests were running stable for 240 days the VFAs could not be fully degraded. The methane yield and the developing of VFA concentrations are shown in Fig. 4 and Fig. 5. The median value of acetic and propionic acid were 1.87 and 4.49 g l^{-1}. This corresponded to the performance of the full scale plant.

The reduction of HRT to 30 (1b) - meaning a higher discharge in the full scale plant – had a significant impact on the Methane yield which was reduced to 148 Nm^3 t_{COD}^{-1}. No further accumulation of VFAs was measured, which indicates process stability on a constantly high VFA concentration level.

The increase of the OLR simulates a future change in substrate composition (4a). A higher COD concentration of 115 g kg^{-1} was achieved by adding starch and lard to the substrate. Although, the methane yield and the COD degradation rate increased in test 4a compared to 1a the additional COD could not be degraded completely. The tests were running stable under similar VFA concentrations and pH. The additional reduction of HRT to 30 days represent a OLR up to 3.85 kg_{COD} m^3 d^{-1} and led to an increase of acetic and propionic acid to 4.50 and 5.21 g l^{-1} respectively, which indicates an overloaded and destabilized process.

3.2.2 Enhancement options of the full scale plant

The COD value is the most reliable indicator for the energy potential of a substrate and stands for the amount of organic compounds, which can theoretically be degraded to methane by the anaerobic microorganisms. The lower the COD content in the final degradation product, the higher is the methane yield and consequently the profitability of a biogas plant.

The options for the full scale plant can be clearly pointed out if the parameter COD reduction, OLR and COD:TKN ratio are set in a context. Fig. 3 illustrates the input parameters COD:TKN ratio and OLR on the horizontal axis. The determining output parameter COD reduction is drawn vertically. The digestion of degradable COD can be optimised by nitrogen reduction to TKN 4 g kg^{-1} at a HRT of 40 days with no additional COD input (Test 3a). All other tested enhancement options for the full scale plant have an impact on COD reduction. A decline of HRT (3b) after removing N had the least influence on COD reduction, but no additional high COD concentrated ABP fractions may be treated in that case.

The implementation of a nitrogen removal unit at the full scale plant would be more economically feasible if a greater volume of ABP could be processed. Therefore the OLR was increased for Stripped attempts to 2.90 and 3.71 $kg_{COD}\,m^3\,d^{-1}$ for 40 (Test 5a) and 30 days (5b) HRT, respectively. The high methane yield of Y 315 $Nm^3\,t_{COD}^{-1}$ at a HRT of 40 days and the reduction of 81.9 % of the degradable COD confirm that greater volume of ABPs can be processed when nitrogen is reduced.

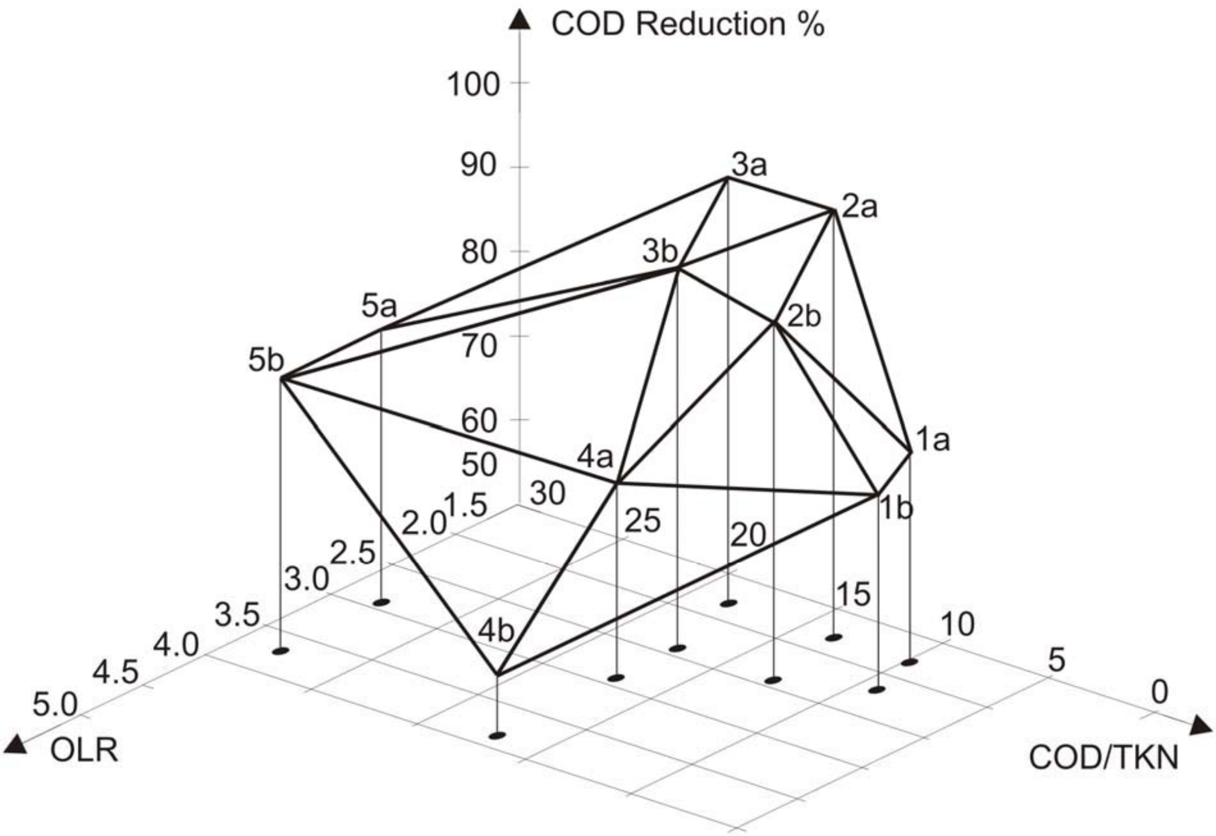

Fig. 3.: Interrelation of set point parameters OLR and COD:TKN with the performance parameter reduction of degradable COD for test attempts Reference and Stripping

Based on satisfactorily COD degradation results a stable digestion process has to be achieved in any enhancement project for the full scale plant. Good parameters for monitoring the process are the concentrations of volatile fatty acids which represent intermediate fermentation products in the anaerobic food chain (Ahring et al., 1995). An accumulation of acids always indicates an imbalance in the microbial interactions or an inhibition by various substances. However, fatty acids are also a potential degradable organic substrate. Therefore, high VFA contents in a fermentation end product mean potential losses in methane yield. In addition, certain levels in the final storage tank of a biogas plant should not be exceeded, as fatty acids have a high potential of causing odour emissions. So, the lower the VFA content at a given OLR or HRT, the better is the degradation performance of a fermenter. In this context Schnürer and Nordberg (2008) suggested from their experiments that digesters to be run on high levels of ammonia should have a HRT longer than 30 days.

3.2.2 Effects of removing nitrogen

The TKN of the inoculum and the substrate was reduced by steam stripping and adjusted to 5.5 and 4.0 g kg^{-1} for experiments 2a, 2b and 3a, 3b, 5a, 5b, respectively. Thus, the COD:TKN ratio was widened to 13 – 18. Instantaneously after the start of experiment 3a the concentration of acetic acid (AA) decreased constantly, whereas propionic acid (PA) slightly increased. The degradation of PA did not start before acetic acid fell below 1.0 g l^{-1} on day 40. Afterwards PA was fully removed within 20 days and stayed under 0.1 g l^{-1} till the end of the experiment after 240 days. The level of AA increased slightly but remains on a very low level of < 1.0 g l^{-1}. (Fig. 5)

After acclimatisation phase of 15 days 100 % of degradable COD or 60.3 % of total COD was removed and a median methane yield of Y 293 Nm^3 t_{COD}^{-1} was achieved (Fig. 4). The influence of nitrogen inhibition was clearly reduced at a TAN concentration of 3.45 g l^{-1}, pH was settled down at a median level of 7.81. Thus, the calculated UAN dropped to 254 mg l^{-1}.

The reduction of HRT by 25 % (Test 3b) did not influence the VFA concentration and pH significantly. It indicates a satisfactorily anaerobic digestion process. The COD degradation and methane yield decreased slightly to 53.0 % (94.6 % of degradable COD) and Y 213 Nm^3 kg_{COD} d^{-1}, respectively.

When the OLR was increased in tests 5a and 5b the partial degradation of VFA showed that a higher ABP input can be applied, but degradation goals might not be fully met. When the HRT was lowered to 30 days (5b) additionally, acetic acid was not degraded any longer and the pH dropped to 7.37. The developing of data is shown in Fig. 4 and Fig. 5. This instability indicates that the limit of organic load (OLR: 3.71) for the bacteria was exceeded.

To clarify the advantage of nitrogen removal, experiments where operated at a TKN set-point of 5.5 g kg^{-1}. Both, acetic and propionic acid were degraded partly following the similar sequence as described above. The remaining concentration of 1.65 to 1.98 g l^{-1} of propionic acid pointed out the existing ammonia inhibition. The calculated UAN of 555 and 542 for tests 2a and 2b reflected the course of pH (7.94 and 7.88) and the TAN concentration of 4.95 and 4.62 g l^{-1}. However, these results showed that even the removal of small portion of nitrogen enhanced the degradation of VFA, but a significant reduction towards TKN 4.0 g.kg^{-1} (or TAN 3.45 g.l^{-1}) is required for any enhancement options to enable a complete degradation of VFAs. (Figure 5). Edström et al. (2003) recommended a TAN concentration of 4 g l-1 for a stable digestion process and reported UAN inhibition (450 - 560 mg l^{-1}) above this limit. Hejnfelt and Angelidaki (2009) found severe process inhibition for TKN concentrations higher than 7 g kg^{-1}

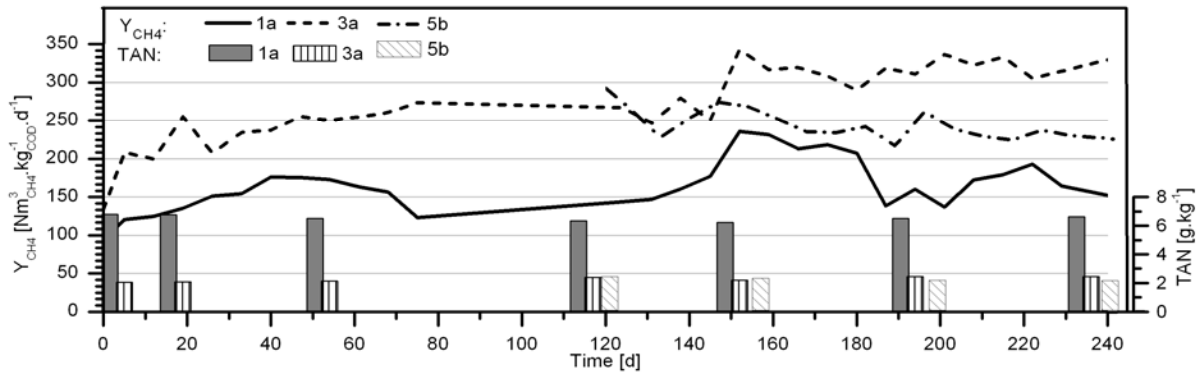

Fig 3.: Methane Yield Y and TAN concentration for Reference test 1a, Stripped test 3a and test Stripped with high OLR and reduced HRT 5b

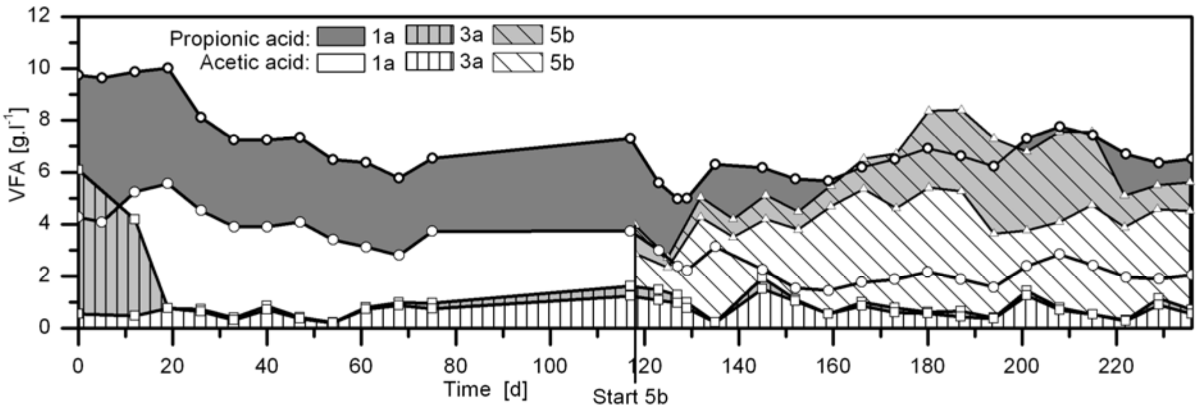

Fig 4.: Developing of acetic and propionic acid concentrations for Reference test 1a, Stripped test 3a and test Stripped with high OLR and reduced HRT 5b over the full time of experiments

3.2.4 Addition of COD

Widening the COD:TKN ratio was conducted by adding COD in terms of corn starch and glycerine to the substrate. Glycerine is a by-product from biodiesel production and easily available for anaerobic digestion bacteria. Accordingly, when having a positive effect on the degradation of the substrate, its methane yield should be highest of all three attempts. In this case the whole COD input coming from glycerine/starch plus a great fraction of the COD input coming from the substrate would be converted into methane. Widening the COD:TKN ratio to 17:1 required an addition of glycerine that increased the OLR to 2.90 kg_{COD} m^{-3} d^{-1} (Test 7). The fast hydrolysis of Glycerine within an inhibited anaerobic consortium led to an accumulation of acetic and propionic acid. Thus, no stable process could be achieved. Changing the COD source to corn Starch (Test 6a) at 40 days HRT a COD reduction of 63.4 % and a methane yield of Y 399 Nm^3 t^{-1} d^{-1} were obtained Although, the digestion continued steadily most of the methane was converted out of the easily available starch and not the waste material which should be treated originally. The high VFA concentrations lowered the pH to 7.68 and consequently lowered the calculated UAN to 318 mg l^{-1}. The reduction of HRT by 25 % (Test 6b) led to an continuing accumulation of VFA and destabilisation of the process. In the Glycerine attempts volatile fatty acids accumulated and even no stable fermentation could be achieved.

Shanmugam and Horan (2009) observed an improved biodegradability – raised from 24.36 to 44.31 % – by increasing the C:N value from 5 to 15 through different mixtures of leather fleshing waste with municipal biowaste. Therefore, with a value of 18.3, the COD:TKN ratio of Stripping and Glycerine should be in an optimal range. Thus, when comparing the results, the question, of whether inhibition is a matter of the ammonia concentration or a matter of the nutrient composition, should get answered.

The results clearly confirm that, the addition of an extra carbon source is no favourable solution to overcome an ammonia inhibition. The extra carbon might be converted, but the but degradation rates of volatile fatty acids is insufficient and the bacterial consortium is still inhibited.

3.2.5 Overcoming ammonia inhibition

The comparison of the degradation performances reveals a significant improvement of the situation after widening the COD:TKN ratio from 9 to 18 through ammonia removal by steam stripping. When taking a closer look at the results of Stripping propionic acid is only being degraded, after the level of acetic acid fell below 1 g l^{-1}. Shortly after the increase of the acetic acid concentration, propionic acid follows. Only Stripping (3a) showed a rapid acetic acid degradation beginning shortly after the start of the degradation experiments (Fig. 5). These results agree well with the literature. Angenent et al. (2002) observed that methane production was mostly channelled through hydrogen after increasing the ammonia level to 3.6 g l^{-1}. Acetate degradation, which still occurred, went probably via a syntrophic relationship between an acetate-oxidizing organism and a hydrogen-utilizing methanogens (Schnürer and Nordberg, 2008).

For halving the specific growth rate, Angelidaki and Ahring (1993) reported a higher sensitivity to ammonia toxicity of the acetotrophic methanogens (3.5 g l^{-1}) than for the hydrogenotrophic methanogens (7 g l^{-1}). Also Koster and Lettinga (1984) found a higher sensitivity of the acetotrophic methanogens when exposed to higher ammonia concentrations. Hence, as approximately 73 % of the carbon flow in anaerobic digestion usually goes via the acetotrophic pathway (Garcia et al., 2000), an inhibition of the acetotrophic methanogens caused by ammonia must have a great impact on the performance of a reactor. Furthermore, propionic acid was found to be a strong inhibitor of the hydrogenotrophic methanogens (de Bok et al., 2003; Wiegant and Zeeman, 1986). This means, an inhibition of the acetotrophic methanogens caused by ammonia leads firstly to an increase of acetic acid. This causes an accumulation of propionic acid which moreover inhibits the hydrogen consumption. Gallert and Winter (2008) confirmed this during the restart of a waste digester and demonstrated to overcome this propionic acid accumulation by re-inoculation.

3.3 Implementation at the full scale plant

The European regulation (EC) No.1774/2002 banned established disposal technologies for ABPs after the pandemic occurrence of different animal diseases. As a result of that and the increasing quantities of non-consumable ABPs the treatment costs rose. Therefore, alternative disposal techniques are pushed forward (European Commission, 2005; Kirchmayr et al., 2007). According to national statistics the anaerobic co-digestion of ABPs in Austria increased constantly from 90,000 tonnes in 2004 to 215,000 tonnes in 2008. Though, there is only one plant situated at a slaughterhouse site exclusively treating ABPs. Although the plant has been operating for 6 years the nitrogen content of the substrate limits any increase of ABP discharge. As anaerobic digestion combines material recovery and energy production it gives a promising solution in both, an economical and ecological view (Salminen et al., 2003).

Nevertheless, anaerobic degradation of protein rich substrates – such as slaughterhouse wastes – leads to high ammonia concentrations in the fermentation sludge (Gallert et al., 1998). This causes problems as unionised ammonia (NH3) is suspected to have a toxic effect on the microorganisms. Anaerobic digestion at high ammonia concentrations leads to an insufficient substrate degradation, reduced methane yields, unstable fermentation conditions and heavy odour emissions at the biogas plant. Hence, if anaerobic digestion should become a competitive treatment technique for nitrogen rich wastes, the ammonia problem has to be solved.

3.3.1 Ammonia removal and recovery

Steam stripping was chosen to remove ammonia directly out of the digester after the first fermentation step. Steam stripping is a simple process that provides high removal efficiency. Rao et al. (2008) outlined that alternative chemical or biological processes to reduce the inhibition by ammonia during anaerobic digestion require significantly more energy and special carbon sources. Hence they coupled a UASB reactor treating poultry manure litter leachate with an ammonia stripper. Another approach assembling a stripping unit in front of an anaerobic digestion process of waste sludge is reported by Nakashimada et al. (2008). Zeng et al. (2006) concluded that because of the high pH value (> 8,0) of fermenter sludges coming from biogas plants that use nitrogen rich substrates, the stripping experiments were done without raising the pH of the substrate. This should on the one hand avoid problems coming from an increased salinity and on the other hand save treatment costs. Stripping the fermenter content without extensive pre-treatment is not feasible using a packed column. Recovering ammonia and closing the ammonia cycle might also be useful for the production of nitrogen fertilizers, which are commonly produced by Haber Bosch synthesis using fossil energy. According to data from USGS (2009) 136 million tonnes were synthesised in 2008 worldwide.

3.3.2 Performance of the digestion after nitrogen removal

The experimental results presented should evaluate the considered extension and enhancement projects. Full scale experiences in Austria and Germany quote a minimum reduction of the volatile dry matter in the range of 80 % as one criterion for reaching a satisfactorily energy efficiency in anaerobic digestion. This value was only achieved by Stripping. Although a stable fermentation process could be achieved in Reference and Starch, its performance is far away from the above stated level.

Thus, clear recommendation for the full scale plant can be derived from the laboratory experiments. The enhancing of the biogas plant will go hand in hand with the future development of the abattoir. A nitrogen removal and recovering unit has to be built at the full scale biogas plant. After implementation and reduction of the TKN concentration to 4 g kg^{-1}, either the HRT in the biogas plant may be lowered by 25 % to 30 days or the OLR may be increased by 61 % to 2.90 $kg_{COD}\ m^3\ d^{-1}$. The first means, that more of the same ABP fractions may be treated. The lift of OLR as second option allows the additional treatment of high concentrated ABPs like pig blood, colon and colon content in the biogas plant. Outlook calculations demonstrate that all on site available ABPs may be treated by the new process. Further volumes after an extension of the abattoir may also be utilized in the anaerobic digestion plant.

4. Conclusions

The experimental results point out, that the enhancement of an anaerobic digestion plant exclusively treating ABPs is possible when the nitrogen content in the process is reduced.

- The reduction from TKN 7.5 to 4.0 g kg^{-1} leads to an increase of COD degradation by 55 % and improves the VFA digestion.
- Thus, in the full scale plant either the OLR of may be increased by 61 % to 2.90 $kg_{COD}\ m^3\ d^{-1}$ or the HRT may be reduced by 25 % to 30 days.
- A satisfactorily mono digestion of all available ABPs requires the removal of nitrogen out of the system.
- A valuable fertilizer is recovered and the nitrogen cycle is closed in an effective way.
- A robust stripping process, that allows the ammonia recovery directly out of the process, has to be developed

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4. Anaerobic digestion of animal by-products

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9

Anaerobic Degradation of Animal By-Products

Roland Kirchmayr, Christoph Resch, Martin Mayer, Stephan Prechtl, Martin Faulstich, Rudolf Braun, and Johann Wimmer

9.1. INTRODUCTION

As a result of growing meat consumption and production the slaughtering and rendering industry has an increasing output of nonconsumable animal by-products. According to new European legislation the established treatment and disposal technologies are either not allowed anymore or are too costly. Therefore, new ways of treatment for raw slaughterhouse waste products and pretreated materials from rendering plants have been established. As legislation on the recovery of organic materials for animal feed is becoming tighter and more restrictive, anaerobic digestion is a promising alternative for the treatment of these materials, since the process combines material recovery and energy production (Salminen and Rintala, 2002). The careless or rash utilization of animal by-products as substrates for anaerobic digestion subsequently may cause process instability and odor emissions. The conditioning of the digestate, like nitrogen-removal or concentration, may reduce logistic costs for utilization as a fertilizer.

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9.2. REQUIREMENTS OF THE EUROPEAN REGULATION EC NO. 1774/2002

9.2.1. Hazard Potential of Animal By-products

The inefficient treatment of slaughterhouse waste or improper use of products produced from it led to the pandemic occurrence of animal diseases such as BSE and foot-and-mouth disease. Only a rigorous European Regulation on the treatment and further use of animal by-products could prevent a further spread of diseases. This very extensive Regulation governs the collection, transport, methods, and procedures of treatment, as well as the further disposal, use, or trade in the products. Therefore, the Regulation has an effect on the export to and the import from nonmember countries of the European community.

9.2.2. Definitions

ABP (animal by-products): Animal by-products are all bodies or parts of animals and products of animal origin not intended for human consumption, because either they are not fit for human consumption or there is no market for them as foodstuff.
Biogas plant according to the ABP-Regulation: This is a plant in which biological degradation of products of animal origin is undertaken under anaerobic conditions for the production and collection of biogas.

9.2.3. The EC-Regulation (EC) No. 1774/2002

The Regulation (EC) No. 1774/2002 of the European Parliament and of the Council of October 3, 2002, lays down health rules concerning animal by-products not intended for human consumption (''ABP-Regulation'' or ''Hygiene Regulation'' in committees working on environmental issues) and regulates possible uses and processing rules of animal by-products (ABP). The Regulation was published in the official journal of the European Community of October 10, 2002, L 273, pages 1–95, and has had to be applied directly in all member states of the European Community since May 1, 2003.

Further temporary Regulations and implementing rules were laid down by the European Commission for the cushioning and modification of this Regulation. In this text all amendments that entered into force by February 2006 are included. Specific possibilities of processing and use for animal by-products are listed in this ABP-Regulation. In future, existing and newly developed methods and processes or utilization possibilities may be examined by the Scientific Steering Committee and approved by the European Commission.

9.2.4. Animal By-products in Biogas Plants

In the ABP-Regulation animal by-products are divided into three categories:

- Category 1: contains those materials with the highest risk for public health, animals, or the environment (hygienic risk, risk of BSE, etc.).
- Category 2: includes all animal by-products that can be allocated neither to category 1 nor to category 3 (e.g., manure or digestive tract content or animals not fit for human consumption). For manure and catering waste the conditions for approval and for treatment, as well as other criteria for the end product and for the remaining animal by-products, are defined (Figure 9.1 and Table 9.5). Biogas plants that process catering waste or manure can be approved by national rules (pending further EC-legislation).
- Category 3: comprises those animal by-products that would be fit for human consumption, but are (for commercial reasons) not intended for human consumption.

According to Regulation (EC) No. 1774/2002 different substrates for biogas plants require different treatments (no treatment, pasteurization, or sterilization) and the approval of the respective biogas plant according to national regulations or regulation (EC) No. 1774/2002. Figure 9.1 and Table 9.1 give an overview about substrates and the legal force of national or international regulations and a survey of materials designated for treatment in biogas plants.

9.2.5. Materials of Category 1

ABP of category 1 represent an increased risk for public health, animals, or the environment. These materials such as specified risk material (SRM), animals suspected of being infected with BSE, ABP with increased concentrations of

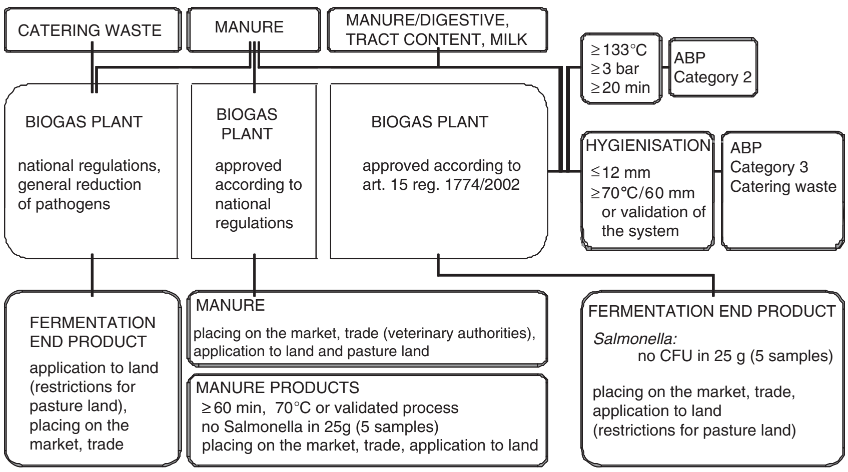

Figure 9.1. Substrates and legal force of national and international regulations.

Table 9.1. Survey of materials designated for treatment in biogas plants: Three categories of animal by-products

Category	Material to be treated in biogas plants
Category 1	Not designated
Category 2 Without preliminary treatment	Manure as well as digestive tract content (separated from the digestive tract; if there is no risk of dispersal of serious-infectious diseases) milk and colostrum
Category 2 After sterilization with steam pressure and marking (with smell)	All materials classified as category 2 (e.g., perished animals or animals slaughtered, but not intended for human consumption)
Category 3 In a biogas plant approved in accordance with article 15 of the regulation	All materials classified as category 3 (e.g., meat-containing wastes from the foodstuff-industry, slaughterhouse wastes of animals fit for human consumption)
Category 3 In biogas plants which are to be approved in accordance with provisions and methods to be adopted or which are approved according to national legislation	Catering waste (except from international means of transport)

Table 9.2. Tissues classified as specific risk material: Regulation (EC) No. 999/2001, amended by Regulation (EC) No. 657/2006

Animal	Tissue
Bovine animals, aged over 12 months	Skull excluding the mandible and including brain and eyes, and the spinal cord.
Bovine animals, aged over 24 months	Vertebral column including the dorsal root ganglia, but excluding vertebrae of the tail, the spinous and transverse processes of the cervical, thoracic and lumbar vertebrae and the median sacral crest and wings of the sacrum.
Bovine animals, all ages	Tonsils, intestines from duodenum to rectum, mesentery
Ovine and caprine animals, aged over 12 months	Skull including brain and eyes, tonsils and spinal cord
Ovine and caprine animals, all ages	Spleen as well as ileum

environmental contaminants, solid materials (> 6 mm) from wastewater treatment in category 1 processing plants and establishments in which SRM is removed (slaughterhouses and cutting plants), and catering waste from international means of transport are not allowed to be processed in a biogas plant.

9.2.6. Materials of Category 2

Category 2 comprises all ABP that are neither included in category 1 nor in category 3. These are manure, digestive tract content, milk not fit for human consumption, killed or fallen animals, and solid materials in wastewater streams of slaughterhouses (particle size > 6 mm). ABP of category 2 may be processed in a biogas plant only after sterilization with steam pressure (at least 20 minutes without interruption at a core temperature of more than 133°C and an absolute steam pressure of not less than 3 bar), except manure, digestive tract content, and milk, which need no pretreatment.

9.2.6.1. Manure

The definition of manure according to the ABP-Regulation is excrements and/or urine of farm animals, with or without litter, and guano, either unprocessed, processed, or transformed in biogas plants or composting plants.

Manure, digestive tract content (separated from the digestive tract), milk, and colostrum are materials of category 2. These materials, however, can be fed directly and without any pre-treatment to an approved biogas plant. The fermentation end product of the ''transformation'' of manure processed in a biogas plant together with other substrates that are not covered by this Regulation (e.g., renewable raw materials or energy crops) may be considered as untreated manure. Conditions for placing ''untreated manure'' on the market within the boundaries of a member state of the EC, as well as special requirements for transport (marking as ''manure,'' cleaning of containers, etc.) may be laid down by national legislation.

The list of third countries from which EC member states may authorize imports of manure for treatment of the soil is established in a separate regulation (Directive 79/542/EEC). Conditions and special requirements for the placing manure on the market within national boundaries may be laid down by national legislation.

9.2.6.2. Wastewater from Slaughterhouses

For slaughterhouses (or cutting plants removing SRM) and plants processing material of category 1 and 2 (e.g., intermediate and rendering plants) a pretreatment of the wastewater is required that retains all solid materials up to a particle size of 6 mm (e.g., using a screen with a mesh size of 6 mm). Any materials removed from the wastewater by this pretreatment unit (screenings, materials from desanding, grease and oil mixtures, sludge, material removed from drains) are regarded as materials of category 2 or materials of category 1 (for plants processing materials of category 1 or removing SRM), respectively.

Any decomposition or reduction in size of the materials in the wastewater stream prior to the retaining unit is not allowed! Materials removed from the wastewater stream after the pretreatment of the wastewater (flotation sludge, etc.) and the residual wastewater containing no solid particles are not covered by this Regulation and are to be treated in accordance with the relevant wastewater legislation.

9.2.7. Materials of Category 3

Category 3 contains all ABP originating from animals fit for slaughter but not intended for human consumption as well as animal by-products from food production and catering waste. Materials of category 3 must be pasteurized before treating in an approved biogas plant. These ABP may be processed in a biogas plant equipped with a hygienization unit which cannot be bypassed. These biogas plants have to be approved according to the approval conditions laid down in article 15 of the ABP-Regulation (see Figure 9.2). The processing standards in the hygienization unit are defined as 70°C during 60 minutes with a maximum particle size of 12 mm. Alternative processes have to be validated in order to demonstrate the achievement of an equal overall risk reduction (Reg (EC) No/208/2006).

9.2.7.1. Catering Waste

Catering waste is grouped into two categories: those from international means of transport (catering from aeroplanes, ships, or railways) are included in category 1 and must be disposed of. All other catering wastes are defined as materials of category 3. Catering waste of category 3 may be processed in biogas plants according to national rules pending the adoption of relevant provisions and approvals of the EC.

Pending adoption of these EC provisions concerning the treatment of catering waste, the application of alternative standards for processing may be authorized for biogas plants processing only catering waste (together with manure as well as energy crops). However, effective reduction of pathogens has to be ensured.

9.2.7.2. Meat and Bone Meal

At present the feeding of animal protein [meat and bone meal (MBM)] to farm animals is prohibited. General exceptions exist for the feeding of animals not intended for human consumption (pets and fur animals). Specific derogations allow the feeding of certain kinds of processed animal protein (i.e., hydrolyzed protein, fish meal) to nonruminants. Furthermore, it is prohibited to feed animals with meat and bone meal produced from bodies or parts of animals of the same species.

9.2.7.3. Fish Waste

Fish waste (category 3) may be fed to a biogas plant passing the hygienization unit or may be processed to microbiologically stable fish silage or compost. The production of fish meal or fish silage for the purpose of feeding farm animals is allowed under certain conditions.

9.2.8. Examples of Animal By-products in Biogas Plants

In the following sections two cases are specified, which shall illustrate the application of the ''ABP-Regulation'' (EC) No. 1774/2002 to biogas plants.

9.2.8.1. Biogas Plant at a Pig Slaughtering Facility

Substrates used in the biogas plant for this case study are listed in Table 9.3. This biogas plant is registered by the competent authorities and has to be approved according to article 15 of the ABP-Regulation. A pasteurization unit that cannot be by-passed must be available and a concept of control and monitoring must be followed. The microbiological parameters for digestates (Table 9.4) must be applied. Manure as well as digestive tract content (separated from the digestive tract) may be processed in the bio-gas plant without pretreatment.

All ABP of category 3 such as slaughtering by-products, bones, intestines, as well as blood are to be pasteurized before processing in a biogas plant. Any materials removed from the wastewater stream before the prescribed wastewater pretreatment unit in slaughterhouses are regarded as materials of category 2 and, like these, are to be sterilized by steam pressure prior to processing in a biogas

Table 9.3. Example for substrates used in a biogas plant located at a pig slaughterhouse

Substrate	Category	Required treatment
Manure from pigs	2	No pretreatment required
Digestive tract content	2	No pretreatment required
Digestive tract (fit for human consumption)	3	Pasteurization
Bones, slaughter by-products	3	Pasteurization
Blood	3	Pasteurization
Parts of slaughtered animals[a] (not fit for human consumption)	3	Pasteurization
Screenings, flotation sludge (> 6 mm)	2	Sterilization
Content of fat removal devices[b] (< 6 mm)	–	
Washings (purely liquid fraction)	–	

[a] From animals fit for human consumption (ante mortem), identified as not fit for human consumption after postmortem inspection.
[b] Without reduction of particle size and removed from the wastewater stream after the wastewater pretreatment unit.

Table 9.4. End product parameters for digestates and fermentation end products

Product	Product parameter
Unprocessed and digested manure Placing on the national market	None
Manure products	Heat treatment (> 60 min., > 70°C) *Salmonella:* absence in 5 samples of 25 g each or validated process treatment for the reduction of creators of spores and toxin, storage in special containers
Fermentation end products Biogas plant approved in compliance with Article 15	
Fermentation end product Manure and catering waste	According to national regulations (with restrictions)

Table 9.5. Example for substrates used in a biogas plant located at a bovine abattoir

Substrate	Category	Required treatment
Manure from bovine animals	2	No pretreatment required
Rumen content (stomach content)	2	No pretreatment required
Rumen	3	Pasteurization
Slaughter by-products, bones	3	Pasteurization
Parts of slaughtered animals[a]	3	Pasteurization
Blood	3 or 1	Pasteurization or incineration
Bones: vertebral column and skull	1	Sterilization, incineration
Intestines	1	Sterilization, incineration
Screenings (bigger than 6 mm)	1	Sterilization, incineration
Content of fat removal devices[b] (particles $<$ 6 mm)	–	
Washings (purely liquid fraction)	–	

[a] From animals fit for human consumption (antemortem), identified as not fit for human consumption after postmortem inspection.
[b] Without reduction of particle size and removed from the wastewater stream after the wastewater pretreatment unit.

plant. Materials of categories 2 and 3 sterilized with steam pressure may be fed to a biogas plant without any further pretreatment. For materials that have passed the wastewater pretreatment unit the ABP-Regulation is not effective.

9.2.8.2. Biogas Plant at a Bovine Slaughtering Facility

Substrates used in the biogas plant for this case study are listed in Table 9.5. The plant is registered by the competent authorities and approved according to article 15 of the ABP-Regulation. Herewith follows the necessity of a pasteurization unit which cannot be bypassed and a concept of control and monitoring for this digestion plant. Additionally, the hygiene parameters for the digestates (requirements on digestates) are to be applied. Manure and rumen content may be fed to the biogas plant without pre-treatment. Rumen, slaughtering by-products, and bones are regarded as ABP material of category 3 fit for human consumption and must be fed to the biogas plant by passing the hygienization unit. If it is guaranteed that no SRM gets into the bloodstream during slaughtering (reliable separation of blood draining unit and SRM removal unit, retention of blood until the submission of a negative BSE test, etc.), bovine blood can be considered as material of category 3. Otherwise, bovine blood is to be ''disposed of'' as a mixture together with SRM.

Materials of category 1 must not be fed to the biogas plant.[1] Content of fat removal devices (achieved after screening with a mesh size of 6 mm) as well as washing (purely liquid fraction) are not subject to any provisions of this Regulation.[2]

[1] At present the biogas technology is not designated for the treatment of material of category 1. It is intended, however, that other methods and processes (other than those mentioned in the ABP regulation) may be assessed by the Scientific Steering Committee (SSC) and approved by the Commission afterward. For that it is required to submit a detailed description of the alternative process method or procedure, including an appropriate risk-assessment, to the Commission of the EC.
[2] They are subject to national and international regulations for wastewater and waste.

9.2.9. Requirements of Fermentation End Products (Digestates)

Depending on the used substrates and the final destination of the digestate different end product parameters are defined in Regulation (EC) No. 1774/2002 (Table 9.4).

9.2.10. Conditions for the Approval of Biogas Plants and Composting Plants According to Article 15 (EC) 1774/2002

Biogas plants processing animal by-products must be registered by the authorities and approved according to article 15 of the ABP-Regulation. Figure 9.2 illustrates which requirements are to be fulfilled in order to be approved by the (competent) authority.

9.2.11. Plants' Own Check / HACCP Concept

The principle of (direct) responsibility of the operator and owner of plants or their representatives is a cornerstone of the demands on the biogas plants on the Regulation (EC) No.1774/2002. By compliance with conditions of acceptance (e.g., nonacceptance of questionable batches) and hygienic operating conditions (e.g., compliance with the ''principle of clean and unclean sector,'' pasteurization

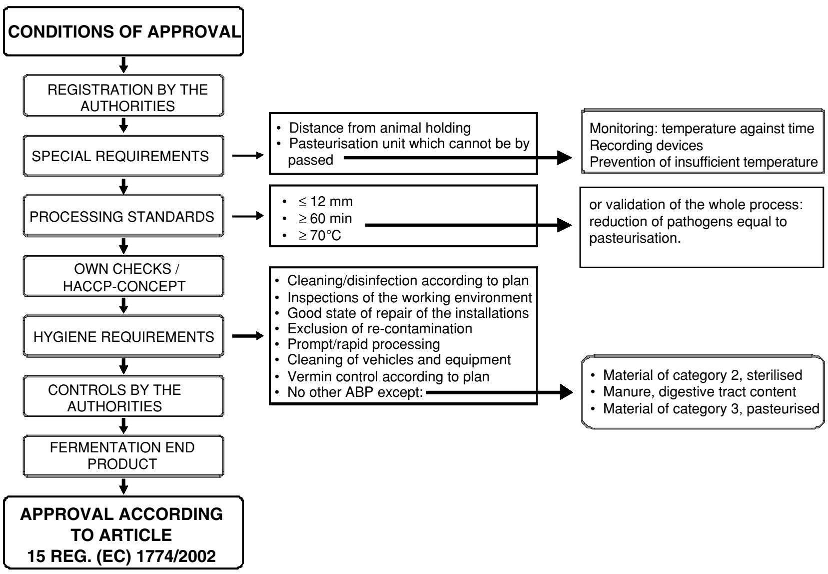

Figure 9.2. Survey of the conditions of approval concerning biogas plants according to article 15 of the ABP Regulation (EC) No. 1774/2002.

of the substrate, etc.) the potential health risk that may emanate from a biogas plant or its digestate can be reduced.

In Regulation (EC) 1774/2002 a concept for self-monitoring according to the system of hazard analysis of critical control points (HACCP concept) is demanded of the operators of biogas plants that use animal by-products as substrates. For biogas plants such a (practicable) concept of control and monitoring that follows the HACCP concept has to be developed.

A description of the critical control points (from a hygienic point of view) when operating a biogas plant must include (Kirchmayr et al., 2003):

- description of the place or procedure that is identified as a critical control point,
- description of the actual risk factors and an evaluation of the significance of these,
- preventive activities in order to minimize or delete the risk,
- monitoring: method and frequency,
- definition of critical values, and
- plan for correcting activities when critical values are exceeded.

Operators of biogas plants have to identify, define, and describe the critical control points of their plants. When operating the plant it has to be controlled and documented whether the parameters or standards in the respective control points have been applied. Examples of control points (from a hygienic point of view) are used when operating a biogas plant and for general hygiene requirements are listed in Tables 9.6 and 9.7.

Table 9.6. Examples for control points in biogas plants (Kirchmayr et al., 2003)

Control points (direct)	Possible failures
Receipt of raw substrate	
Manure	High contamination with pathogens
Vegetable substrates	Unintentional pathogen contamination
Animal substrate	Insufficient pretreatment
Material of category 2 and/or 3; pretreated or not pretreated	
Transport	Mixture of untreated raw material and end product (treated)
Separation between raw material and end product	
Animal by-products	
Hygienization	Insufficient hygienization
Pasteurization material category 3	Failure in temperature, time, particle size Exceptionally high pathogen contamination
Storage of the end products	Recontamination from environment Growth of remaining pathogens
Delivery	Cross-contamination between clean and unclean areas, means of transport and containers
Transport of heat-treated material or end product	

Table 9.7. Examples for general hygiene requirements (Kirchmayr et al., 2003)

General hygiene requirements	Possible failures
Maintainance of premises and equipment	Generally dissatisfactory hygiene conditions
Intercalibration and check of monitoring and measuring instruments	Insufficient validity of the control
Prevention of spreading animal diseases	Insufficient separation and distance from animal holdings and feed storage facilities
Pest control	Spreading of pathogens
	Cross-contamination between clean and unclean sector
Cleaning and disinfection	Internal reinfection (''in-house'')
	Continuous contamination of heat-treated substrates
	Cross-contamination between clean and unclean sectors (i.e., via aerosols)
Education of staff	Uncertainty about aim and methods of the plant's own check system and unclear delegation of responsibility

9.3. CHARACTERIZATION OF WASTE STREAMS

9.3.1. Slaughterhouse Wastes

The physical and chemical characteristics of slaughterhouse wastes and pretreated rendering wastes are extensively reported in literature. Analyses of chemical parameters and characteristics have been done for a lot of reasons and different treatment goals. In recent years the focus of waste analyses focused also on methane potential investigations to evaluate the feasibility of these materials for treatment in anaerobic digestion plants. These investigations started on slaughterhouse and rendering wastewaters and were extended to solid organic waste fractions.

Based on these results anaerobic digestion plants have been set up on slaughterhouse and rendering sites, most of them connected to the wastewater treatment facilities. Under the new Regulations (EC 1774/2002) more and different waste fractions may be treated in anaerobic digestion plants. Some of these waste fractions contain high amounts of nitrogen which has to be incorporated in feasibility assessments. In Table 9.8 analysis of waste fraction and raw by-products that accrue directly at the slaughterhouse site are listed. The analysis data are cited from literature or unpublished analysis conducted at own laboratories.

In modern slaughterhouses most waste streams are collected separately. Therefore best substrate mixture for the anaerobic digestion plant may be designed. As this ''best substrate'' cannot be achieved in lot of cases, all listed chemical parameters have to be taken into account when the substrates are designed:

- The COD or VSS contents are used to calculate the derived hydraulic and organic loading rate.
- The TKN load for the anaerobic digester is the result of the nitrogen contents of chosen substrate composition.

Table 9.8. Substrate characteristics from waste fractions at slaughterhouses

Substrate	COD (g/kg)	TKN (g/kg)	TS (%)	VSS (%)	Biogas or methane potential	Reference
Swine wastes						
Gut fill (swine)		1.57	11.2	9.9	2,040 Nm^3_{Gas}/Mg_{VSS}	Carpentier et al. (2005)
Swine blood		17.07	14.8	14.3	1,020 Nm^3_{Gas}/Mg_{VSS}	Carpentier et al. (2005)
Swine blood	340	33.13	21.48	20.44		Unpublished results: Authors' own investigations
Swine colon	259	1.27	6.12	5.43		
Swine gut	299			12.08		
Swine stomach content	215	2.96	13.53	13.03		
Bovine and cattle wastes						
Bovine blood	310	21.17	19.48	18.63		Unpublished results: Authors' own investigations
Bovine leg bones	1,181	24.89	76.13	50.01	522 Nm^3_{Gas}/Mg_{COD} 221 Nm^3_{Gas}/Mg_{VSS}	
Bovine clutches	547	49.16	47.59	32.83	527 Nm^3_{Gas}/Mg_{COD} 317 Nm^3_{Gas}/Mg_{VSS}	
Bovine shoulder bones	707	36.46	69.26	40.70	330 Nm^3_{Gas}/Mg_{COD} 191 Nm^3_{Gas}/Mg_{VSS}	
Bovine rib bones	770	32.53	58.28	39.51	819 Nm^3_{Gas}/Mg_{VSS} 422 Nm^3_{Gas}/Mg_{VSS}	
Bovine omasum	646	14.01	29.59	27.48		Unpublished results: Authors' own investigations
Bovine omasum content	188	6.05	11.87	10.93		
Bovine rumen	119	2.17	9.55	8.43		
Bovine stomach content	166	11.94	10.24	10.03		
Poultry wastes						
Poultry blood	273	10.83	10.59	9.47	878 $Nm^3_{CH_4}/Mg_{VSS}$ 305 $Nm^3_{CH_4}/Mg_{COD}$	
Poultry blood		1.67	22.00	20.02	500 $Nm^3_{CH_4}/Mg_{VSS}$ 100 $Nm^3_{CH_4}/Mg_{FS}$	Salminen et al. (2003)
Poultry carcass			37.00		200–250 $Nm^3_{CH_4}/Mg_{FS}$	Chen (1999); Chen and Shyu (1998)
Poultry litter		1.67–2.99	52–81	32–53	140–220 $Nm^3_{CH_4}/Mg_{VSS}$ 100–150 $Nm^3_{CH_4}/Mg_{FS}$	Webb and Hawkes (1985)
Poultry manure		0.92–3.15	20–47	12–31	200–300 $Nm^3_{CH_4}/Mg_{VSS}$ 40–60 $Nm^3_{CH_4}/Mg_{FS}$	Huang and Shih (1981); Safley et al. (1987)
Poultry feather		3.65	24.30	23.50	200 $Nm^3_{CH_4}/Mg_{VSS}$ 50 $Nm^3_{CH_4}/Mg_{FS}$	Salminen et al. (2003)
Poultry offal, feet, head		2.07	39.00	37.05	700–900 $Nm^3_{CH_4}/Mg_{VSS}$ 300 $Nm^3_{CH_4}/Mg_{FS}$	Salminen et al. (2003)
Poultry trimmings and bone		3.44	22.40	15.23	600–700 $Nm^3_{CH_4}/Mg_{VSS}$ 150–170 $Nm^3_{CH_4}/Mg_{FS}$	Salminen et al. (2003)

The composed substrate has to fulfill both: to provide the organic load and to guarantee a nitrogen concentration within the reactor that is not close to the inhibitory level. The highest nitrogen concentrations have been investigated in the blood of cattle, swine, and poultry.

If bones or carcasses are included in the substrate, the high inorganic part may be the limiting step for the use of these waste fraction. As these bone fragments will sediment in the reactor a process strategy has to be implemented. The gas and methane yield for several investigated bones of cattle and bovine varies widely. Thus the meat that is stuck to the bone depends on the cutting and separation process in the slaughterhouse. The investigated cattle leg bone gave a high gas yield ($522\,Nm^3_{gas}/Mg_{COD}$) because of the marrow within the bone.

9.3.2. Waste Fractions from Rendering Plants and Food Processing Industries

Twenty-five percent of the total weight of slaughtered animals have to be processed according to the regulations for treatment and use and disposal of these slaughtering by-products (Tritt and Schuchardt, 1992). Lots of these wastes as carcasses and blood are processed in rendering plants to output basically market products, solid organic wastes, and wastewater. Some of the market products are not allowed to be used any longer and also have become waste products that have to be disposed. The currently installed processes that transform slaughterhouse waste and animal by-products into new wastes will not be cost and energy effective in some future cases. Other technologies such as anaerobic digestion may be implemented into existing rendering facilities to dispose animal by-products accomplishing following goals:

- hygienization,
- recovery of valuable products,
- recovery of energy, and
- purification of wastewater.

A series of substrate analysis, laboratory, and pilot-scale experiments (Tritt, 1990; Johns, 1995; Kirchmayr, 1998; Huber, 1998) have been undertaken to assess the feasibility of implementing anaerobic digestion technology into rendering plants. The substrate characteristics and measured biogas yields are listed in Table 9.9. In many cases the available wastes in rendering and slaughterhouses do not have constant chemical characteristics. The parameters vary because of changed or inconstant processes. The waste fractions showing the widest variations are the fat sludges from dissolved air flotation (DAF) and filter-cakes from sieving machines. Peak discharges during washing cycles may overload the flotation or sieving units and cause significant changes in the characteristics of the flotation sludges and sieving residues.

The final process step in a rendering plant will be a wastewater treatment unit that includes an aerobic nitrification and denitrification step. This has to be taken

Table 9.9. Substrate characteristics from waste fractions at rendering plants and various wastes from meat processing industry

Substrate	COD (g/kg)	TKN (g/kg)	TS (%)	VSS (%)	Biogas[a] or methane potential; substrate specification	Reference
Waste fraction from rendering plant						
Homogenate	975	24.40	42.70	39.40	1 140 Nm^3_{Gas}/Mg_{VSS}	Kirchmayr (1998) laboratory and 4 m^3 pilot scale
Blood	141	15.6	9.70	9.20	630 Nm^3_{Gas}/Mg_{VSS}	
Concentrated blood	21.2	3.95	1.5	1.0	940 Nm^3_{Gas}/Mg_{VSS}	
Stomach and intestinal content	205	6.41	16.5	13.7	680 Nm^3_{Gas}/Mg_{VSS}	
Rumen content	185	3.78	14.3	12.7	370 Nm^3_{Gas}/Mg_{VSS}	
Exhaust vapors condensate	15	0.55	0.05	0.05	Clear, bright yellow liquid	Huber (1998) laboratory and 4 m^3 pilot scale
Concentrated blood	17	2.23	1.33	0.8	Turbid liquid	
Blood and wastewater from the hutches	54	5.34	4.99	4.06	Viscous, coagulated blood residual	
Homogenate	705	18.18	36.25	33.19	Viscous, pasty	
Concentrated sludge	114	5.94	7.64	6.87	Viscous up to gel-like	
Squeezed fat sludge	1,596	15.54	88.47	81.4	From fluid to solid, depending on temperature	
Substrate mixture[b]	71	2.64	3.10	2.76	500–700 Nm^3_{Gas}/Mg_{COD} CH_4: Ø 73% of biogas	
Wastes sludges from sieves and fat flotation						
Primary flotation sludge			16.00	15.00	2,400 Nm^3/Mg_{VSS}	Carpentier et al. 2005, laboratory and 2 m^3 pilot scale
Filter-cake from sieving		9.15	31.50	30.50	1,000 Nm^3/Mg_{VSS}	
Floated fat after DAF	100	1.27	6.12	5.43		Unpublished results: Authors' own investigations
Fat scraper content (cattle slaughter-house)	214	3.09	14.98	11.39		

Byproducts from meat processing industry					
Squeezed stomach from swine	347	18.89	15.40	14.03	750 Nm^3_{Gas}/Mg_{VSS} 304 Nm^3_{Gas}/Mg_{COD} 709 Nm^3_{CH4}/Mg_{VSS} 287 Nm^3_{CH4}/Mg_{COD}	Unpublished results: Authors' own investigations laboratory scale
Concentrated animal fat and proteins	511	2,720	30.14	26.46	607 Nm^3_{Gas}/Mg_{VSS} 376 Nm^3_{Gas}/Mg_{COD} 314 $Nm^3_{CH_4}/Mg_{VSS}$ 195 $Nm^3_{CH_4}7Mg_{COD}$	
Category 3 material (swine)		16.29	38.5	37.8	1,670 Nm^3_{Gas}/Mg_{VSS}	Carpentier et al. (2005), laboratory and 2 m^3 pilot scale
Animal glue leather from tannery industry	217	8.71	17.10	10.69	430 $Nm^3_{CH_4}/Mg_{COD}$	Grasmug (2004), laboratory scale
Carcass meal from rendering plant	1,101	82.70	95.77	64.58		
Mixed waste 50% poultry blood; 50% OFMSW	263	8.56	11.35	8.90	60 $Nm^3_{CH_4}/Mg_{FS}$ CH_4: Ø 68% of biogas	Resch et al. (2005), 4 m^3 pilot scale
Mixed waste 30% conc. animal fat and proteins 70% OFMSW	330	12.17	19.81	16.12	102 $Nm^3_{CH_4}/Mg_{FS}$ CH_4: Ø 72% of biogas	

[a] Best results concerning gas production and stability were achieved with a hydraulic retention time (HRT) of 40 days and a volume load (B_R) of 1.5 kg $COD/m^3 \cdot d$, respectively.
[b] Composition of mixture: exhaust vapors condensate 76.1%, concentrated blood 10.0%, homogenate 6.6%, blood and wastewater from the hutches 4.1%, sewage sludge 2.7%, fat sludge 0.5%.

into account when the substrate input for the anaerobic digestion plant is composed of available waste fractions. The denitrification process step in the wastewater treatment unit needs an easy bioavailable carbon source to transform nitrate and nitrite into N_2. A waste fraction consisting of easy bioavailable COD and low nitrogen concentration may be used for this purpose. Exhaust vapors condensates from the rendering process achieve these requirements.

The anaerobic digestion technology is widely used in organic waste treatment from households [organic fraction of municipal solid waste (OFMSW)], industrial, and catering facilities (Mata-Alvarez, 2000). Treatment plants that are licensed to accept the mentioned category 3 wastes also may use category 3 animal by-products from slaughterhouses or meat processing industries. As some fractions of animal by products may cause severe problems in the biological treatment process as single substrate the co-digestion with other waste streams may be a favorable option (Grasmug, 2004). The OFMSW is an easily degradable material with low nitrogen contents and therefore is a highly suitable cosubstrate for animal by-products and wastes from meat industry. Results from laboratory and pilot experiments are listed in Table 9.9.

9.4. TECHNICAL SCALE EXPERIENCES WITH THE ANAEROBIC DIGESTION OF SLAUGHTERHOUSE WASTES

Since the ABP-Regulation (EC No. 1774/2002) is defining new treatment possibilities and laying down corresponding processing parameters, new pathways for the utilization of slaughterhouse wastes were opened. Some biogas plants in the agroindustrial sector are using pasteurized ABPs as cosubstrates (together with manure, catering waste, and other energy crops). There is only fragmented information available about the limits of the utilization and the behavior of high-protein-containing wastes for anaerobic digestion in laboratory, pilot, and technical scale (Brachtl, 2000; Carpentier et al., 2005; Resch et al., 2005; Kirchmayr, 1998; Grasmug, 2004).

9.4.1. Anaerobic Digestion of Slaughterhouse Waste in an Agricultural Biogas Plant

In the following, experiences with a full-scale plant using animal by-products deriving from a pig slaughtering facility processing 16–20 t/d of slaughterhouse wastes will be described. Colon, colon content, blood, and fat scrubber content, stomach content, and process water are used as substrates. The plant originally was designed and constructed like a ''classic'' agricultural biogas plant. Two main fermenter (HRT = 100 d in fermenter 1, HRT = 50 d in fermenter 2) and a post-fermentation tank (without heating, HRT = 40 d) were constructed. The digestate was pasteurized (70°C/60 min) after the anaerobic digestion and subsequently stored in the storage tank. The digestate was used as fertilizer.

Table 9.10. Characteristic parameters of the sludge in the fermenter of a biogas plant (monitoring interval: 1 year)

Sample		pH	TSS (%)	VSS (mg/L)	TKN (mg/L)	NH_4-N (mg/L)	UAN 37°C (mg/L)
Fermenter 1	min	7.60	2.59	1.94	6,668	5,867	333
	max	8.25	3.33	2.67	8,172	7,069	1,154
Fermenter 2	min	7.87	2.49	1.87	7,488	6,494	518
	max	8.30	2.75	2.12	8,093	7,120	1,269
Postfermentation	min	7.96	2.10	1.55	7,125	6,115	588
	max	8.31	2.44	1.81	7,944	6,972	1,179
Storage tank	min	7.89	1.51	1.04	6,120	5,300	581
	max	8.56	4.81	3.16	7,742	7,584	1,782

Due to a start-up without monitoring, several serious process failures were observed. Hand in hand with process instability, heavy foaming together with the accidental escape of fermentation liquid occurred. The subsequent land application of the (not completely metabolized) digestate contributed to the range of increased odor emissions.

In order to ensure proper operation, the biogas plant concept was redesigned. In section 6 the concept to reduce odor emissions deriving from the processing unit is described.

Previous to anaerobic digestion, the substrate is pasteurized. With this measure the anaerobic digestion will be situated at the ''clean side'' of the process (concerning the requirements of the ABP-Regulation) and the manipulation of the sludge is much easier.

Before feeding into fermenter 1 and fermenter 2 (in parallel) the substrate is further chilled down to 50°C in order to minimize damage of the bacterial biomass in the biogas fermenter. Both fermenters are operated at mesophilic temperatures (35°C). Higher temperatures will increase the amount of unionized ammonia (NH_3, UAN) in the system. Table 9.10 shows the calculated UAN concentrations in the specific biogas plant. Levels between 300 and 1,700 mg/liter are close or beyond reported inhibition values (Koster, 1986; Koster and Koomen, 1988; Angelidaki and Ahring, 1994). Calculated on the basis of Eqs. 1 and 2 (Lide, 1993) the increase in temperature will cause an increase in unionized ammonia concentration by 43% (at pH = 8). This in turn could probably lead to a much higher level of inhibition.

According to the high water content of the substrate mixture the total nitrogen content in the fermenter is in the range of 5.8 to 7.5 g/liter. In agricultural cofermentation plants using maize corn as main substrate, ammonia values in the same range have been measured (IFA-Tulln, 2005) apparently without any toxic influence.

$$(1) \qquad \frac{[NH_3]}{[NH_3] + [NH_4^+]} = \left[\frac{1}{1 + 10^{pKa-pH}}\right]$$

$$(2) \qquad pKa = 4 * 10^{-8} * T^3 + 9 * 10^{-5} * T^2 - 0{,}0356 * T + 10{,}072$$

The applied organic loading rate was limited to a maximum of 3.5 kgCOD/m^{3}*d. Laboratory-scale experiments to investigate the biodegradability of slaughterhouse wastes under anaerobic conditions showed that an organic loading rate exceeding 2.5 kgCOD/m^{3}.d may cause process stability problems (Kirchmayr et al., 2002).

After reinoculation and operation at low organic loading rate the process is quite stable, although the concentrations of free volatile fatty acids are very high (see Table 9.11). The concentrations of acetic and propionic acid are reduced in the secondary fermenter and storage tank from 5000 to 9000 mg/liter to approx. 1000 to 2000 mg/liter. The organic loading rate may not be considered as very high, but as shown in Section 6.2 the end product of the anaerobic digestion may still cause some annoyances due to odor emissions.

Considering the reasons for the incomplete metabolization only little information is available and different hypotheses about the toxic effect of intermediate metabolites have been described. On the one hand the high concentration of unionized ammonia may cause toxic effects (Koster, 1986; Koster and Koomen, 1988; Angelidaki and Ahring, 1994), on the other hand different intermediate metabolites like phenol, p-cresol, or indole also may cause inhibitory effects (Behmel et al., 1994; Field et al., 1987; Griehl et al., 2002). Higher concentrations of volatile fatty acids also may be identified as the source of inhibitory effects (Kroeker et al., 1979; Duarte et al. 1982; Ahring, 1995).

Further minor process instabilities may be explained as a result of elevated concentrations of cleaning and disinfecting agents contained in the fat scrubber fraction. Also, a partial overload of the flotation device may have caused process instability due to a slight hydraulic overload of the anaerobic reactors.

9.4.2. Conclusion

Under the described circumstances the stable operation of a biogas plant using only slaughterhouse waste as substrate is feasible. Strong attention should be paid to the

Table 9.11. Volatile fatty acid concentrations in the biogas reactors with slaughterhouse waste as substrates (monitoring interval: 1 year)

Sample		Acetic acid (mg/L)	Propionic acid (mg/L)	i-Butyric acid (mg/L)	Butyric acid (mg/L)	Valeric acid (mg/L)	i-Valeric acid (mg/L)	VFA total (g/L)	UVA[a] total (mg/L)
Fermenter 1	min	5,196	3,539	339	104	893	60	11.71	3.35
	max	9,451	6,235	1,213	500	2,361	578	19.04	23.35
Fermenter 2	min	2,784	4,240	172	0	331	52	8.70	2.53
	max	7,832	6,425	1,006	268	1,997	458	16.06	9.25
Postfermentation	min	775	1,130	39	0	36	0	4.34	1.54
	max	4,682	6,294	529	111	819	162	10.36	4.59
Storage tank	min	409	140	24	0	17	16	0.98	0.28
	max	3,370	3,141	228	112	923	179	3.55	2.94

[a] Calculated as acetic acid.

odor emissions caused by the operation of the biogas plant and by the land application of the digestate. To ensure higher metabolization rates during anaerobic digestion and in order to utilize the microbial activity in the storage tank for further stabilization of the digestate the hygienization unit (pasteurization) should be located before anaerobic digestion and not afterwards.

The anaerobic monodigestion of high nitrogen-containing animal by-products like slaughterhouse wastes causes a very weak equilibrium in the process. Small mistakes may easily cause process failure. Proper training of the plant operator and careful process control (e.g., trough the measurement of volatile fatty acids, ammonia content, and pH) is absolutely necessary in order to ensure long-term digestion operation. Further investigation is necessary to determine which process parameters have to be changed (e.g., nitrogen removal in a bypass stream) in order to increase the reported insufficient degradation rate.

9.4.3. ATZ Thermal Pressure Hydrolysis (TPH) of Animal By-products

In this section the application of ATZ thermal pressure hydrolysis (TPH) followed by anaerobic digestion is described (Prechtl et al., 2002, 2003). The TPH process with subsequent anaerobic digestion has been so far successfully explored concerning the recycling of sewage sludge (Hertle and Renner, 1994; Tippmer, 1994; Chwistek et al., 1997) and organic fraction of sorted municipal solid waste (ATZ Entwicklungszentrum, 1997). The sterilization in the TPH reactor (temperature about 230°C, pressure 20–30 bar) results in the splitting of the organic polymers by hydrolysis with the guarantee of a complete hygienization.

After mashing with process water the hydrolysate is converted into biogas in a fixed-bed-loop-reactor. The biogas is used to supply the thermal energy for the TPH process and for NH_3-stripping.

Due to good degradation performance in the anaerobic step there is only a small amount of solid residue left, which may easily be dewatered and disposed through incineration. Compared to the existing rendering plants less wastewater is produced due to the process water cycle and the loss of water through the step of NH_3-stripping and biogas production. The excess wastewater has a typical composition like a normal wastewater from anaerobic waste fermentation plants, with COD concentrations between 4 to 7 kg/m^3 after conditioning.

After pretreatment with the TPH process it is possible to run a stable monofermentation of animal by-products with a high organic loading rate of about 10 kg $COD/m^{3*}d$. According to the methane content of up to 77% in the biogas and a biogas yield between 200 to 300 m^3/mg raw material, a combined heat and power station produces up to 780 kWh electric energy from 1,000 kg of raw material.

An anaerobic biofilm reactor with a volume of 85 m^3 was used for biogas production from the hydrolysate produced in the TPH pretreatment. With this process a reduction of 80–94% $COD_{dissolved}$ could be achieved. Figure 9.3 shows a flow diagram of the industrial-scale application.

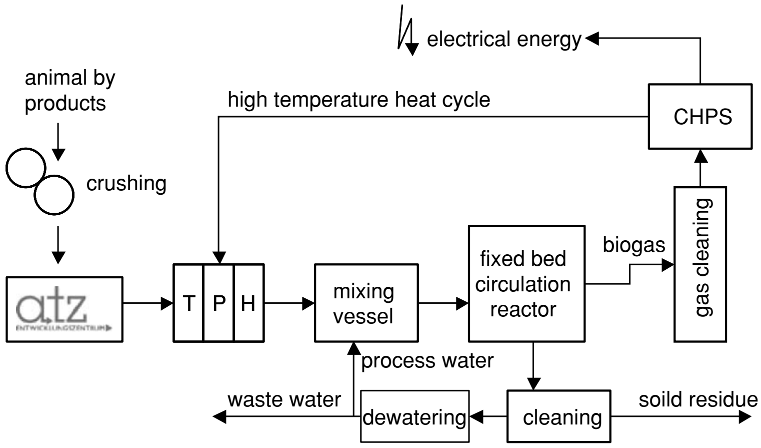

Figure 9.3. Flow diagram of the process combination ATZ-TPH and digestion of animal by-products.

Contrary to the present production of meat and bone meal, which consumes fossil energy, the process combination of TPH and anaerobic digestion is able to produce renewable energy from animal by-products. The biogas generated from the organic share of the animal by-products by fermentation can be used to produce an excess of thermal and electric energy for the whole process.

Since May 2005 a full-scale TPH plant, with a capacity of about 40,000 mg/year, for the treatment of manure and renewable substrates is in operation in Germany. The plant was constructed by the R. Scheuchl company, a licensee of ATZ Entwicklungszentrum.

9.5. NITROGEN REMOVAL IN THE ANAEROBIC DIGESTION

Anaerobic digestion effluents and its subsequent handling now are gaining more interest not only because plant sizes are growing significantly, but also because of rising environmental interest and politics, and thus implementation of stricter limits throughout the European Union. But effluent purification also is interesting in terms of on-site liquid recycling. Many industrial wastes have to be diluted, suspended, pulped, etc., in order to condition the waste stream for anaerobic digestion. As some slaughterhouse waste streams have extremely high nitrogen contents dilution is a proper way to lower the nitrogen concentration in the reactor (Siegrist et al., 2005). Actually, the following treatment strategies are presently implemented, whereas agricultural use is a final depose, the other technologies provide regenerated water applicable for further use on-site:

- Agricultural use as fertilizer
- Biological treatment

- Magnesium ammonium phosphate precipitation (MAP)
- Ammonia stripping

In the following section the treatment concepts will be described briefly and the effluent quality after treatment will be outlined.

9.5.1. Agricultural Use as Fertilizer

Agricultural use of digestion effluent is the only treatment approach where the whole stream, including the liquid and the solid fraction, is disposed. In some European countries this is a common approach, regulated by the amount of nitrogen applied to the field. Thus the council Directive 91/676/EEC is limiting the use of nitrogen fertilizer to 170 kg N/ha/a. In terms of costs this solution can be a very cheap one, but disadvantages are that huge storage vessels have to be built and the use is laborious. For larger anaerobic digestion plants or for industrial plants handling large volumes, agricultural use will not pose a solution. Particularly for larger amounts of digester effluent, fertilizer use is not appropriate as the corresponding demand on the agricultural area will be enormous. Along with the rising demand for agricultural area, the transportation costs will rise dramatically.

9.5.2. Biological Nutrient Removal

Biological nutrient removal by means of carbon, nitrogen, and phosphor removal (Obaja et al., 2003) is a well know and common practice at least when municipal wastewater is treated (Verstraete and Philips, 1998). Treatment of ammonium-rich wastewater in industrial application on the other hand is still under way. One challenge is the potentially inhibitory characteristics of ammonium-rich wastewater on the biological process (Brond and Sund, 1994), another is the poor carbon to nitrogen ratio (Ra et al., 2000).

As far as the inhibitory effects (Anthonisen et al., 1976) are concerned a smart treatment concept will overcome these difficulties. Only the initial break in phase attention has to focus on the special wastewater property. With a smooth start-up high loaded industrial wastewater also can be treated, even more so as activated sludge systems are well adaptable.

For denitrification an organic carbon source is mandatory. A poor carbon to nitrogen ratio therefore will result in reduced denitrification (Elefsiniotis et al., 2004). With optimal nitrification an accumulation of nitrate or nitrite will take place. If a lack in carbon source is identified, treatment concepts are proposed using external carbon source. Such systems are realized in advanced biological treatment mainly following the sequencing batch reactor (SBR) concept (Vallés-Morales et al., 2004).

All together these processes comprise two steps of treatment (Carrera et al., 2003). The first step is oxidation of the ammonium to nitrite or nitrate followed by denitrification.

The two steps of biological nitrification (Pambrun et al., 2004):

(3) Nitritation: $NH_4^+ + 1.5\ O_2 \rightarrow NO_2^- + 2H^+ + H_2O$

ammonia-oxidizing bacteria

(4) Nitratation: $NO_2^- + 0.5\ O_2 \rightarrow NO_3^-$

nitrite-oxidizing bacteria

Subsequent to nitrification, denitrification takes place. During an anoxic period, nitrite and nitrate are used as an oxygen source. The N_2 builds up and is released to the environment. Thus the total nitrogen concentration in the anaerobic digester effluent is reduced, making the liquid ready for recycling.

Recently, apart form classic denitrification anammox plants (Fux et al., 2002), anaerobic ammonium oxidation also is applied. Thereby ammonium is converted to nitrogen gas under anoxic conditions with nitrite as the electron acceptor (Mulder et al., 1995).

9.5.3. Magnesium Ammonium Phosphate Precipitation (MAP)

Crystallization of ammonium and phosphate through MAP from anaerobic digestion effluents is implemented not only for removing these nutrients, but also for recovering them (Doyle and Parsons, 2002; Uludag-Demirer et al., 2005; Battistoni et al., 2000). Hence, this not only contributes to minimizing the environmental impacts but also contributes to the increasing global demand for nitrogenous fertilizer (Mulder, 2003) and the limited phosphorus rock reserves (Steen, 1998). Struvite precipitation has already been applied to various wastewaters such as swine waste (Burns et al., 2001), tanning factories (Tunay et al., 1997), and anaerobic supernatant (Yoshino et al., 2003) and follows the general and simplified reaction outlined in Eq. 5.

(5) $$Mg^{2+} + PO_4^{3-} + NH_4^+ \Leftrightarrow MgNH_4PO_4 * 6H_2O$$

For Mg^{2+} various sources can be used, like $Mg(OH)_2$, MgO, $MgCl_2 * 6H_2O$, and others. Depending on the initial molar ratio of Mg:N:P (Altinbas et al., 2002) an addition of a phosphorus source also may be necessary. For optimal results the pH has to be adjusted (Nelson et al., 2003), but this results in a rise in salinity. This elevated salinity has to be considered when opting for an internal recycling of the liquid. As far as final discharge is concerned, a subsequent treatment is obligatory in order to meet the discharge quality as the total carbon load needs to be considered.

9.5.4. Ammonium Stripping

Prior to stripping of ammonium the suspended solid concentration should be lowered and the pH of the supernatant has to be increased. The higher the pH, the more the equilibrium of ammonium to ammonia is shifted toward NH_3. That is necessary for economical operation of the stripper. After this pretreatment step, free ammonia (NH_3) is stripped off by air or steam (Siegrist, 1996).

Stripping of volatile compounds is a standard industrial application, but stripping anaerobic digester effluent needs further care. The high suspended solids concentration especially has to be considered. Like MAP precipitation, ammonium stripping barely affects the total COD load, and therefore not sufficient for direct wastewater discharge. But in terms of internal recycling of the liquid, this technology is highly suitable and furthermore a valuable product, ammonium concentrate, can be obtained and sold. Similarly to MAP precipitation, ammonium stripping is only possible with the usage of chemicals resulting in a significant rise in salinity.

9.5.5. Purification Grade

The purification grade not only depends on the chosen technology but also on the quality of the substrate used for anaerobic digestion (Figure 9.4).

As the quality as well as the quantity of digester effluent varies tremendously from plant to plant (Graja and Wilderer, 2001) there is no first-rate treatment technology. Depending on the desired quality, different technologies have to be chosen and in some cases even a combination of them will be beneficial. In Figure 9.4 the average effluent qualities are outlined. COD is hardly affected by MAP precipitation and ammonium stripping, total nitrogen removal is in the range of 80 to 90%. Biological treatment without the use of an external carbon source will

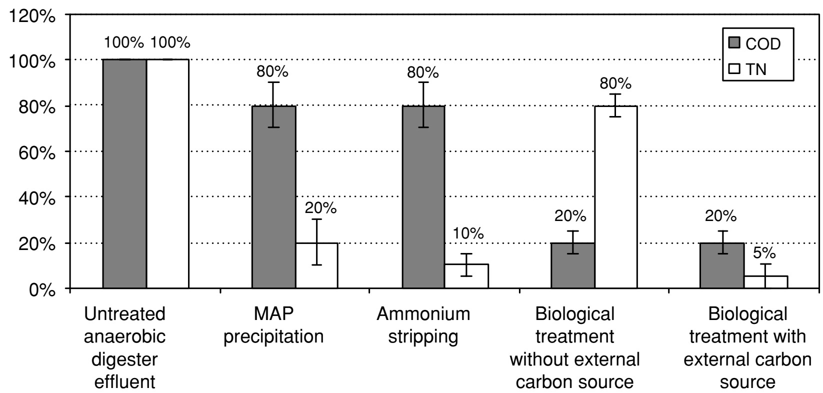

Figure 9.4. Effluent qualities after different treatment technologies by means of COD and TN, calculated on the basis of untreated anaerobic digester effluent. The different treatment options account for different purification levels. The figure shows that COD is nearly unaffected by MAP precipitation and ammonium stripping. Best result can be achieved with biological technologies (Kabdasli et al., 2000; Graja and Wilderer, 2001).

largely eliminate the COD, but the nitrogen content is not significantly affected (Sharma and Ahlert, 1977). Biological treatment with external carbon source results in up to 100% removal on the condition that the activated sludge is totally removed.

9.5.6. Conclusion

On a case-by-case basis the optimal solution will be either a single-treatment process or a combination of several ones. For internal recycling of the liquid for dilution, suspending, pulping, etc., chemical–physical but even more so biological treatment approaches are suitable. For final discharge only a biological treatment meets the legal limits.

Moreover, the ''end of the pipe'' approach will not always be the best, especially when dealing with high nitrogen-loaded waste streams like slaughterhouse wastes. Particularly unionized ammonium nitrogen (UAN) will create problems during anaerobic fermentation, as UAN is inhibits the biological processes. Thus ammonium removal already in the input material will have some beneficial effects: higher loading rates of the anaerobic digester, more stable biological processes, and effluent with a dramatically reduced nitrogen load, simplifying further treatment steps.

The implementation of the stripping procedure in full scale prior to the anaerobic digestion is not reported, but research work is ongoing. Figure 9.5

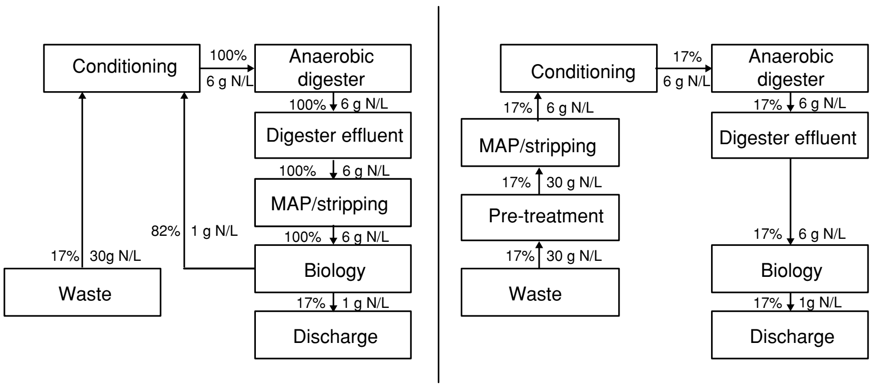

Figure 9.5. The left flow schema shows a treatment concept with postnitrogen removal. The right flow schema shows a treatment concept with prenitrogen removal. The % values indicate the mass flow, with the appropriate nitrogen concentration in grams nitrogen per liter. The difference between post- and pretreatment is that with pretreatment only 17% with high nitrogen content has to be treated, whereas with posttreatment 100% with a relatively low concentration need treatment. Thus the installed treatment capacity has to be accordingly larger. In both systems the nitrogen concentration in the anaerobic digester is 6 g N/L, in the left schema by 5.8-fold dilution with cleaned effluent, in the right schema by nitrogen removal by MAP/stripping.

shows the different flow schemas. In the input material the greater part of the nitrogen is organically bound to the substrate matrix. Hence prior to the nitrogen removal process an adequate pretreatment process has to be installed, decomposing the organic matrix and releasing the nitrogen as ammonium. A further benefit when removing the nitrogen load from the input material is that the high loaded waste streams can be treated. Hence the removal will work more efficiently as the concentration difference is higher, resulting in a reduction of costs.

9.6. ODOR EMISSION

Several biogas plants processing animal by-products such as slaughterhouse wastes and catering waste are facing opposition in residential areas due to massive odor emissions. In the following, the concept to prevent odor emissions deriving from the processing unit is presented. Odor emissions do not only derive from the processing unit, but also the digestate used as fertilizer can cause annoyances.

9.6.1. Odor Emissions from the Processing Unit

The following explanations are focused on the description of the odor emissions from a biogas plant processing slaughterhouse wastes such as intestines, blood, and fat scrubber content. Beside extensive problems concerning the technical process (in the first stage stable fermentation could not be achieved), massive and extensive odor problems occurred already in the start-up phase. The first redevelopment (reduction of the organic loading rate, treatment of the waste air deriving from the central storage tank, and an outdoor storage tank with container biofilters) did not result in a reduction of the complaints about odor emissions.

Olfactometric emission measurements showed extremely high concentrations of odorous substances and loads especially in the central storage tank. The concentration of H_2S (up to approx. 160 ppm) and organic sulfur compounds like dimethyl sulfide, dimethyl disulfide, etc. (approx.$50\,mg/m^3$) in the raw waste air of the pump storage and the central storage tank lead to distinctive acidification of the biofilter materials and further to insufficient deodorization (Figure 9.6). Similarly also the diffuse odor emissions deriving from the gas engine cooling air caused by unsuitable sealing were noticeable. The emissions coming from the open storage tank could not be quantified. Estimation on laboratory-scale (adjustment of equation) showed a high potential for odor emissions.

A reclamation concept developed on the basis of the described measurements (Figure 9.7) specified the separated collection of all highly concentrated exhaust air streams and their usage as combustion air for the gas engines after alkaline washing. Reduced charged waste air streams will be collected and treated in available biofilter systems. To avoid further emissions of the open storage tank the liquid surface was covered with shredded straw.

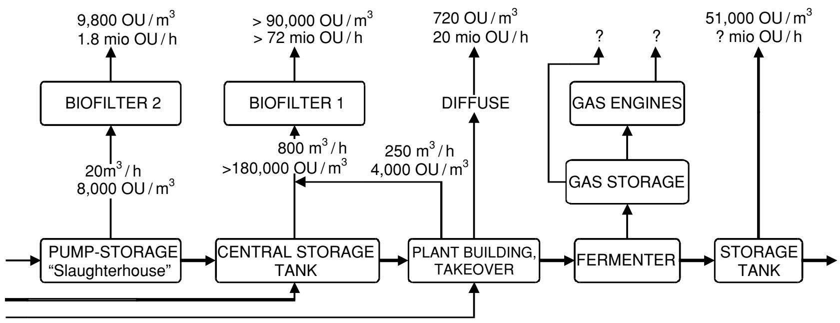

Figure 9.6. Olfactory emissions after the first reclamation trial.

Subsequent checkups confirmed the success of the reclamation strategy. High odorous substance loads coming from the central storage tank and hygienization tank can be effectively reduced by using the gas engines for thermal treatment of the exhaust air. Nitrogen oxide aromas dominate the olfactory odor tone of the exhaust air coming from the gas engines. Raw gas smells were not detectable any more.

The application of heavily loaded waste air streams is not assumed to essentially influence the odor emissions of the gas engines. The estimated odor emissions of the gas motors are remarkably high, probably due to high NO_x emissions of more than 2000 mg/m^3 (remark: Austrian emission threshold value: 400 mg/m^3NO_2). At the time of recording the data both biofilters were operating with approx. 35% of their capacity (in order to generate good degradation performance).

The main causes of annoyance should now be the emissions from the open storage tank and the manipulation of the digestate. Thus subsequently covering the storage tank (reclamation step 2) seems necessary.

To avoid widespread annoyance in a residential area the operation of biogas plants using slaughterhouse wastes as substrates can only be achieved by:

- Storing the substrate and digestate in closed containers, which are connected to the gas system (storage tank) or ventilated sufficiently with subsequent proper treatment of the waste air.
- The emerging waste air including the waste air from hygienization and pasteurization facilities should not be treated in biofilter systems because of the high content of inorganic and organic sulfur compounds. The best solution seems to be to utilize the waste air as combustion air in the existing gas engines (after pretreatment in alkaline scrubbers). As experience shows the engine operation is not noticeably affected as long as the waste air fraction does not exceed 50% of the combustion air.
- Performing all other takeover and manipulation activities (including manipulation of the digestate) in closed, sufficiently ventilated buildings (target

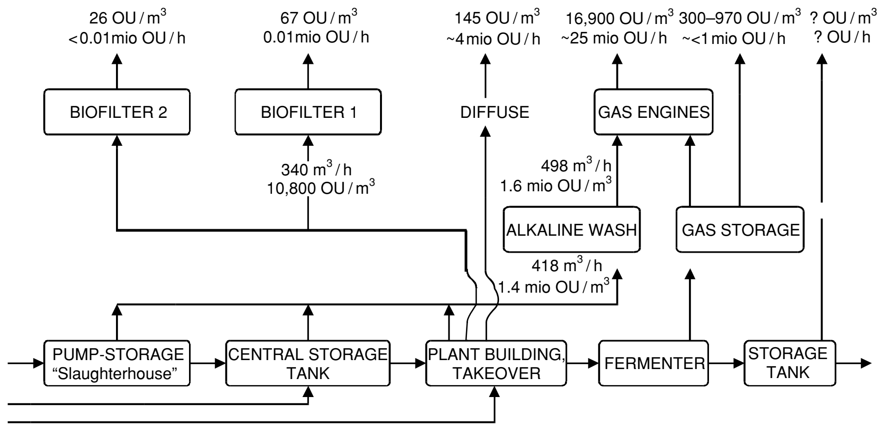

Figure 9.7. Olfactory emissions after implementation of reclamation step 1.

air change number 2–5 to prevent diffuse olfactory emissions). The waste air from the manipulation hall is less odorous, and thus a treatment in biofilter systems seems to be adequate.

- In general all activities that lead to surface contamination with malodorous substances should be minimized (immediate removing of puddles, frequent cleaning, etc.).

9.6.2. Odor Emission during Land Application of Slaughterhouse Digestates

Odor emissions are not only reported from the processing unit, the land application also may result in noticeable odor annoyances. In order to understand land application annoyances two aspects have to be considered: the odor emission concentrations and the hedonic odor tone, i.e., its position on the ''pleasant–unpleasant scale.''

Figure 9.8 gives an overview of the odor emissions observed during test applications of manure and some biogas digestates. The odor emissions of digested slaughterhouse wastes (in this particular case good mineralized, VFA concentrations below 1 g/liter) are comparable with the odor emission deriving from pig and cattle manure (Table 9.12 and Figure 9.8). In this test series only the emissions from incomplete digested catering waste (VFA concentration approx. 13 g/liter) are worse.

Comparing the hedonic odor tone of the different effluents of biogas plants (Table 9.13) it appears that the smell of the digestate of slaughterhouse waste is characterized as very disagreeable (remark: in this case: 7.9 g/liter VFA). Three out of four persons tested characterized the smell as galling and one as very disagreeable. The same result also may be obtained with pig manure. Although

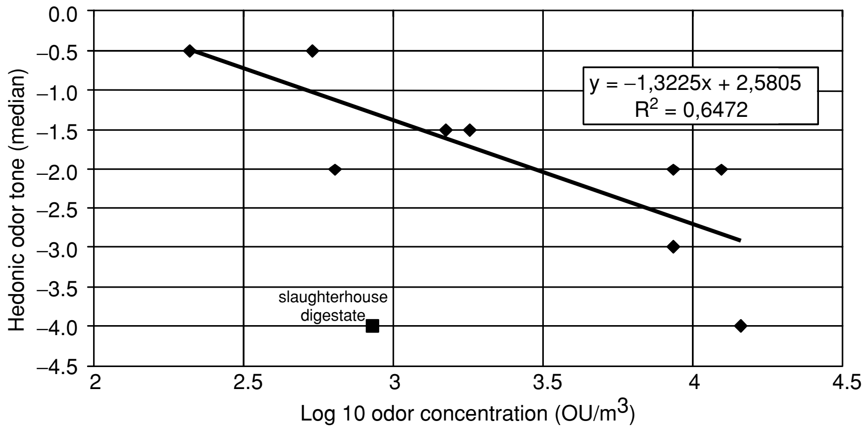

Figure 9.8. Dependence of hedonic odor tone and dilution of different manures and biogas digestates (Weber–Fechner law).

Table 9.12. Soil application of odorous effluents of biogas plants (gas collection: 16.2 $m^3/m^2 * h$)

Substrate	Time after application (soil incorporation 4 h after application) 2	6	24	72	168	Applied digestate[a] (m^3/ha)
	Odor concentration air over soil (OU/m^3)					
Cattle manure	94	30	59	34	34	29.4
Pig manure	164	38	33	20	43	30.3
Effluent slaughterhouse waste	118	16	25	22	22	16.4
Effluent catering waste	750	64	31	31	25	14.3
Effluent energy crops	49	26	29	22	19	32.3

[a] According to a single application of approx. 100 kg N per ha.

the digestate (not well digested) of catering waste is emitting high amounts of odor units (Table 9.12) the smell is not characterized as that offensive.

Generally the experiments show a correlation between odor concentration and hedonic odor tone following Weber–Fechner law except the slaughterhouse digestate produced a very unpleasant odor characteristic at comparably low odor concentrations.

Odor emissions deriving from good mineralized slaughterhouse waste should not exceed odor emissions of pig manure during land application. With a weak reactor performance (corresponding with high VFA concentrations) a much higher level of annoyance deriving from the digestate cannot be excluded.

Table 9.13. Hedonic characterization of biogas plant effluents. Four persons nominating the corresponding hedonic characterization

	Hedonic characterization									
	galling			not pleasant and not unpleasant					delightful	Odor conc.
	−4	−3	−2	−1	0	1	2	3	4	(OU/m^3)
Pig manure	3	1								14,500
Pig manure II			3	1						12,400
Cattle manure				2	1	1				210
Effluent slaughterhouse waste	3	1								850
Cofermentation I		1	1	1	1					1,800
Cofermentation II		2		2						8,600
Energy crops + manure				2	2					540
Cofermentation III		1	3							640
Cofermentation IV		3	1							8,600
Catering waste		2	2							1,500

9.7. ABBREVIATIONS

ABP	Animal by-products
BSE	Bovine spongiform encephalopathy (mad cow disease)
Cat.	Category
CFU	Colony forming units (number of bacteria)
COD	Chemical oxygen demand
conc.	Concentration
DAF	Dissolved air flotation
EC	European Community
FS	Fresh substance
HRT	Hydraulic retention time
MAP	Magnesium ammonium phosphate
OFSMW	Organic fraction of municipal solid waste
TKN	Total Kjeldahl nitrogen
TSE	Transmissible spongiform encephalopathy (e.g. BSE)
TSS	Total suspended solids
VFA	Volatile fatty acids
VSS	Volatile suspended solids

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Commission Regulation (EC) No 808/2003 of May 12, 2003 amending Regulation (EC) No 1774/2002 of the European Parliament and of the Council laying down health rules concerning animal by-products not intended for human consumption (OJ L 117/1,13/05/2003 P.0001–0009).

Commission Regulation (EC) No 811/2003 of May 12, 2003 implementing Regulation (EC) No 1774/2002 of the European Parliament and of the Council as regards the intra-species recycling ban for fish, the burial and burning of animal by-products and certain transitional measures (OJ L 117/14,13/05/2003 P.0014–0018).

Commission Regulation (EC) No 197/2006 of 3 February 2006 on transitional measures under Regulation (EC) No 1774/2002 as regards the collection, transport, treatment, use and disposal of former foodstuffs (OJ L 32, 4.2.2006, P. 13–14).

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5. The impact of increasing energy crop addition on process performance and residual methane potential in anaerobic digestion

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The impact of increasing energy crop addition on process performance and residual methane potential in anaerobic digestion

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Abstract In a full-scale agricultural biogas plant, the changes in process performance connected with the increasing energy crop addition were monitored. The substrates applied were pig manure, solid energy crops and agricultural residues. During the study, the organic loading rate and the volume-related biogas productivity were doubled to 4.2 kg VS/(m^3d) and 2.83 Nm^3/(m^3·d) respectively, by means of increasing the energy crop ratio in the feedstock to 96.5% (volatile solids). This resulted in an increase of the electrical capacity on a level twice as high as before. At the same time, methane yield and organic degradation rate decreased slightly to 0.35 Nm^3/kg VS_{added} and 87.4%, respectively. The strongest impact observed was on the transfer of partly degraded organic material into the digestate storage and with this, an increase of the residual methane potential of the digestate. A maximum theoretical methane load in the digestate of 14.4% related to total methane production of the biogas plant was observed. This maximum level could be reduced to 5.5%.

Keywords Agricultural biogas plants; co-digestion; digestate quality; energy crops; organic loading rate

Introduction

The co-digestion of biomass and sewage sludge or manure is a simple way to increase biogas productivity of anaerobic digesters, especially in isolated rural zones. There are mainly two approaches to implementing biomass addition: to enhance biogas production in an existing digester or to substitute annual feedstock fluctuations. In both approaches, the aim is to run a biogas plant more economically. However, as shown in the present study, those changes in the feedstock and/or the organic loading rate may influence digester performance also in a negative way. However, there is little information available on the consequences and the limits of energy crop addition into biogas plants which have been operated on liquid substrates such as sewage sludge or manure. The major part of the available data have been generated in laboratory-scale experiments. In 1980 Stewart had however published results of laboratory digestion experiments with silage of energy crops (Stewart, 1980) and Jarvis *et al.* carried out some studies in 1997 with grass clover silage (Jarvis *et al.*, 1997), althought the co-digestion of manure and energy crops did not come into focus until recent years. Angelidaki *et al.* (2005); Lehtomäki (2006) and Nordberg *et al.* (2007) presented experimental results on process performance and degradation efficiency applying different loading rates of energy crops. Furthermore, Lehtomäki carried out experiments on different energy crop ratios in the substrate mix, but whereas she stopped her experiments at a energy crop ratio of 40% (VS), in the present study a maximum ratio of 96.5% was applied. As a result of the former lack of information, in most biogas plants process parameters such as the

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substrate mix and the organic loading rate (OLR) were mainly based on proven experiences from existing plants. Data from the monitoring of agricultural full-scale plants presented by Weiland *et al.* (2004); Laaber *et al.* (2005) and Lehtomäki (2006) reflect this development. However, first positive experiences with the enhancement of already existing biogas plants, as in the present plant, showed very good results. Now, more and more operators are attempting to copy this development but ignoring the negative aspects.

The main objective of the present study was to investigate the impact of a change in the feedstock towards energy-rich substrates such as grains of maize or other cereals and the enhancement of the OLR on the digestion process. As well as the upper limits of an OLR increase, consequences in the process or in efficiency parameters such as the biogas yield or the organic degrading rate connected with the process enhancement should be retained. During the study, the electrical capacity of the plant was doubled, therefore some additional parts had to be integrated into the existing plant as is shown in Figure 1.

Material and methods

Full-scale equipment

The biogas plant where this study was realised is situated at a farm in Lower Austria. The plant started as a two-stage continuous stirred tank reactor (CSTR) with a main digester volume of 2,000 m^3 (D1), a second digester step of 1,850 m^3 (D2) and open effluent storage tanks of 3,800 m^3. The average process temperature was 39 °C. The substrates applied during the present study were pig manure (VS approximately 4% VS) and solid energy crops: 37% silage from maize (approximately 30–35% VS), 49% ground grains from maize and wheat (approximately 65% VS) and 14% residues from vegetable and sugar processing (approximately 10% VS). The main part of the process enhancement was achieved by the dosage of silage from maize grains into D2. Biogas was used to generate electrical current and heat. In the beginning one combined heat and power unit (CHP) with an electrical capacity of 500 kW and a thermal capacity of 586 kW (GE Jenbacher, Austria) was run. Electricity was fed into the national grid and heat was used for a local heat supply in the neighbouring village. In the context of the capacity enhancement, a substrate dosage into D2 and a second combined heat and power unit with the same capacity as the first one were installed (Figure 1). These modifications resulted in a total electrical capacity of 1 MW and a thermal capacity of 1,172 MW.

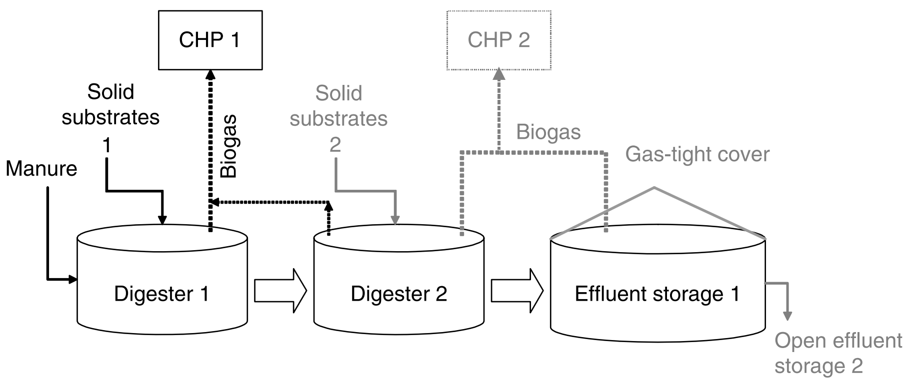

Figure 1 Schematic of the studied biogas plant. Additional parts are pictured in grey

The operation of the studied agricultural biogas plant could be separated into three phases:

Phase 1: Operation as a two-step 500 kW biogas plant with a main and a second digester and a final uncovered digestate storage tank; dosage of substrates only into digester 1; energy crop ratio in the substrate mix: 50% of fresh matter (FM), 92.5% of volatile solids (VS).

Phase 2: Transition phase to 1 MW electrical capacity; dosage of substrates into both digester steps; final uncovered digestate storage tank; energy crop ratio in the substrate mix: 66% of FM, 96% of VS.

Phase 3: Final status: two main digesters followed by a first covered and final uncovered digestate storage tanks; energy crop ratio in the substrate mix: 66% of FM, 96% of VS.

Performance parameters of the plant and chemical parameters of the digesters and the digestate storage were recorded for each phase separately as average values of 6 months (phases 1 and 2) or 12 months (phase 3).

Sampling and chemical analysis

Samples of the digester content were taken every second week, in D1 out of a valve directly in the digester wall and in D2 out of a recirculation loop. The digestate storage was sampled monthly through a valve in the vertical part of the overflow tube out of D2 into the effluent storage. The sampling procedure always consisted of discarding the old content of the sampling tube, taking a sample of 15 L into a bucket and then taking a quota of 0.5 L out of the bucket. The samples were cooled down to 4 °C immediately after sampling. Volatile fatty acids (VFA) were analysed using high performance liquid chromatography (Agilent 1100, RI-Detector). Total nitrogen (TKN), total ammonia nitrogen (TAN), pH, total solids (TS) and volatile solids (VS) were determined using standard methods (APHA, 1998).

Residual methane potential

The residual methane potential in the digestate was evaluated through anaerobic batch fermentation tests. These tests were carried out according to the modified norm DEV S6, DIN 38 414-S6 (DEV, 2005). Thereby, 500 mL of the sample was incubated at 35 °C. Every batch fermentation test was carried out in three replicates. The experiments were continued until no more gas production was observed.

Calculation of performance parameters

The hydraulic retention time (HRT) was calculated according to Helffrich (2005) as a quotient of the daily feedstock mass (fresh matter; FM) and the total digester volume. The organic loading rate (OLR) is the quotient of daily mass of volatile solids (VS) in the feedstock and the volume of the liquid digester content. The organic degradation rate characterises the substrate degradation efficiency and was calculated in a mass balance between substrate input, effluent and biogas production. It is expressed as a percentage [%] of the VS-input. The volume-related biogas productivity is the quotient of the volume of daily biogas production and total digester volume and was used to assess digester efficiency.

Results and discussion

Process monitoring

Performance parameters. The development of the performance parameters was mainly influenced by the doubling of the electrical capacity of the plant maintaining the same digester volume at the beginning of phase 2. Figure 2 shows the development of the electrical capacity and the volume-related biogas productivity. At the beginning of phase 3, a stable performance on the maximum electrical capacity was reached. The OLR also stabilised at twice the level compared to phase 1, after a maximum level of 5.5 kg VS/(m^3·d) in phase 2. According to the studies of Weiland *et al.* (2004) and Laaber *et al.* (2005), which describe the performance of full-scale biogas plants in Germany and Austria, the applied OLR in the studied plant was higher than in the majority of the biogas plants. In Austria the median OLR was 3.39 kg VS/(m^3·d) in 2004/2005 (Laaber *et al.*, 2005).

At the same time, biogas yield, methane yield and power yield, all of them related to the organic loading of volatile solids, returned almost to former levels after a temporary decrease in phase 2 (Table 1). A permanent decrease in methane yield, as was observed by Lehtomäki (2006) after the increase of the OLR from 2 to 3 kg VS/(m^3·d), could not be observed. The specific methane yield decreased during phase 2 and recovered to a level 10.3% below at 0.35 Nm^3 CH_4/kg VS_{added}. In comparison to data from Lehtomäki (2006) and Nordberg *et al.* (2007), who described 0.27 Nm^3 CH_4/kg VS_{added}, and Jarvis *et al.* (1997) who observed 0.30 Nm^3 CH_4/kg VS_{added} in the digestion of energy crops, the observed methane yield in this study was higher, even during process enhancement. This indicates a long-term adaptation of the microbial population to the feedstock.

While Lehtomäki (2006) observed a decrease in the methane yield already at an energy crop ratio higher than 40% in the co-digestion with manure, the data in this study show that an energy crop ratio of 96.5 (VS) in the feedstock was feasible without any decrease.

One of the most important parameters describing plant efficiency is the organic degradation rate. In phase 1 an average level of 89.7% was observed. As a consequence of the change from a two-step to a one-step process and the strong increase of the energy crop-OLR in phase 2, the degradation rate decreased to 82.8%. Similar observations were made by Andersson and Björnsson (2002), who recorded decreases in COD reduction of 10–20% after increasing the OLR in different digester designs. In the final status with

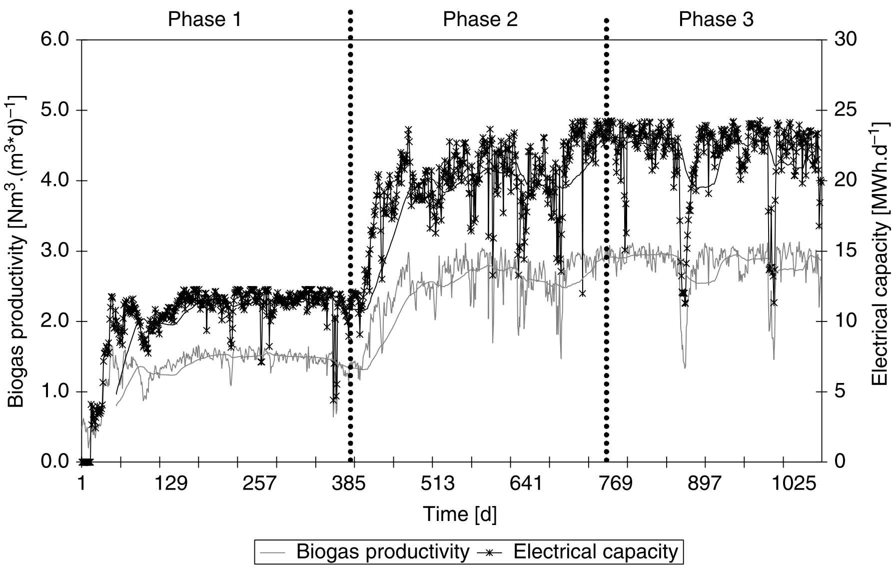

Figure 2 Electrical capacity of the plant and volume-related biogas productivity. Trendlines were calculated as an average of 50 days

Table 1 Development of process parameters

		Phase 1*		Phase 2*		Phase 3**	
Input solids (FM)	[t/d]	14.05	± 1.89	32.58	± 2.76	29.35	± 3.69
Input manure	[t/d]	15.0	–	17.5	–	17.5	–
Input solids (VS)	[t/d]	7.55	± 1.11	16.85	± 1.93	15.14	± 1.99
Input VS (total)	[t/d]	8.10	± 1.07	17.64	± 1.81	16.07	± 2.07
Biogas production	[Nm^3/d]	5755	± 446	10280	± 1137	10908	± 1169
Methane production	[Nm^3/d]	3132	± 246	5154	± 614	5601	± 633
Biogas yield Y_{VS}	[Nm^3/kg VS]	0.72	± 0.11	0.59	± 0.08	0.68	± 0.08
Methane yield Y_{VS}	[Nm^3/kg VS]	0.39	± 0.06	0.29	± 0.05	0.35	± 0.04
Biogas productivity	[$Nm^3/(m^3 \cdot d)$]	1.49	± 0.12	2.67	± 0.30	2.83	± 0.30
Production of power	[MWh/d]	11.42	± 1.00	20.52	± 2.38	22.03	± 2.61
OLR	[kg VS/($m^3 \cdot d$)]	2.10	± 0.28	4.58	± 0.47	4.17	± 0.54
HRT	[d]	133	–	77	–	82	–
Degradation rate	[%]	89.7	–	*82.8*	–	87.4	–

* Average of 6 months.
** Average of 12 months.

the covered digestate storage, a degradation rate of 87.4% was calculated. The differences between phases 2 and 3 shown in Table 1, can be traced back to the covering of the first digestate storage which lengthened the total effective HRT in the gas tight biogas system from 82 to 132 days.

Chemical parameters in the digesters. Chemical parameters inside the digester represent the composition of the substrate mix. As can be seen in Figure 3, three to four hydraulic retention times are needed to map changes in the substrate mix and to establish a new chemical balance. As a consequence of the increasing solid substrate addition at the beginning of phase 2, chemical parameters inside the digester changed significantly. In D2 these changes between phase 1 and phases 2/3 were even larger than in D1, as there was no direct dosage of substrates at all in phase 1. After the installation of the direct substrate dosage into D2 the contents of VS and TS, as well as TKN and TAN, levelled off at a balance, regulated by the contents of these parameters in the substrate mix and the digestate of D1. As in D1 most values in phase 3 returned to levels between phase 1 and

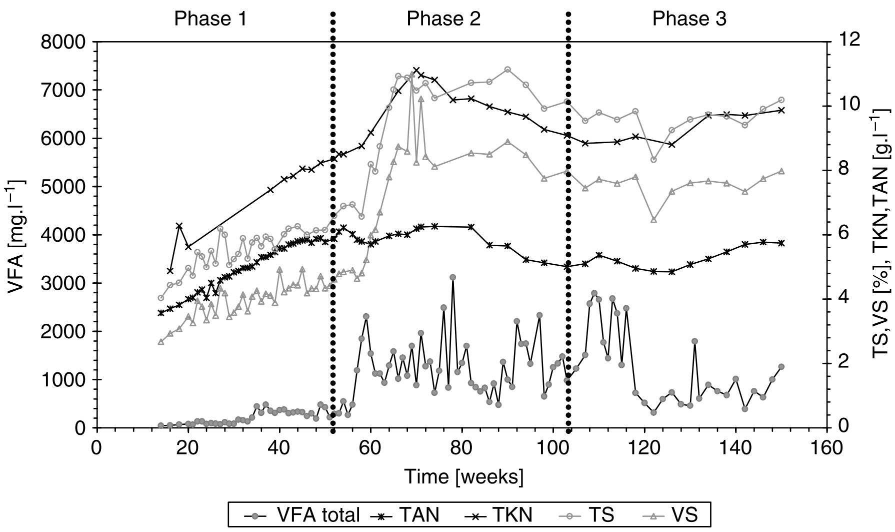

Figure 3 Dynamic of chemical parameters in D2

phase 2. In Figure 3 the development of the chemical parameters in D2 during the different phases is shown.

Generally, it can be summarised that after a transition phase during the enhancement of the biogas plant from 500 to 1,000 kW (phase 2), in phase 3 a stable balance between bacterial biomass and substrate dosage was established again. As in phase 1 the process was limited by substrate dosage as can be seen in the low concentrations of VFA.

Chemical parameters of the digestate. As in D2, the transition from phase 1 to phases 2/3 had a very strong impact on the chemical parameters of the digestate. In phase 1 the theoretical HRT of substrates in the digesters was 133 days. With the installation of the substrate dosage at D2 and the higher total OLR at the beginning of phase 2 the HRT in D2 was reduced to approximately 40 days and the total HRT of the plant to 77 days. These changes resulted in a strong increase of TS, VS and COD in the digestate due to an increasing transfer of partly degraded material into the digestate storage (Table 2 and Figure 5).

Residual methane potential

One of the critical parameters that changed considerably during the different phases, was the residual methane potential (RMP) in the digestate. The RMP is strongly connected to the organic load which is transferred into the digestate. In Figure 4 the daily VS load into the digestate storage, calculated on the VS content and the HRT, in comparison to the OLR, of the plant is shown. As can be seen, in phase 2 the organic load increased to a level three times higher than in phase 1, whereas the VS content and the OLR only doubled. A maximum daily VS discharge of 2.8–3.25 t was found. This corresponds to 17–19% of the daily substrate input. In phase 1 this was 11.2%. As a consequence of covering the first digestate storage the daily discharge of VS was reduced to 2.25 t, which corresponds to 14.0% of the input. It is very difficult to measure the real methane production from the digestate, as the temperature inside the digestate storage is mostly unknown. In the studied plant the average temperature inside the storage was 28 °C depending on liquid level and season. To compare results with other studies the RMPs were calculated on the maximum achievable methane yield determined in batch fermentation tests at 35 °C. Figure 5 shows the methane yields obtained from the digestate at 35 °C at different sampling dates. In addition, one test was carried out at 28 °C to represent the real situation in biogas plants.

Table 2 Development of the chemical parameters of the digestate during three years. Phase 3 represents the current status

		Phase 1*	Phase 2*	Phase 3 **	Phase 3** Maximum	Phase 3** Minimum
pH	[]	7.83	7.79	7.75	7.84	7.66
COD	[mg·L^{-1}]	57.0	119.7	103.9	117.9	79.0
TS	[%]	5.43	10.37	9.51	9.85	9.18
VS	[%]	3.87	8.18	7.48	7.72	7.14
TKN	[g·L^{-1}]	6.50	9.35	9.33	9.79	8.53
TAN	[g·L^{-1}]	4.65	5.90	5.34	5.76	4.88
FAN (37 °C)	[mg·L^{-1}]	396	511	375	486	295
Acetic acid	[mg·L^{-1}]	158	529	662	869	368
Propionic acid	[mg·L^{-1}]	31	83	81	186	26
Total VFA	[mg·L^{-1}]	230	661	772	1063	394
TAN in TKN	[%]	71.5	63.1	57.2	–	–
VS in TS	[%]	71.3	78.9	78.7	–	–

* Average of 6 months.
** Average of 12 months.

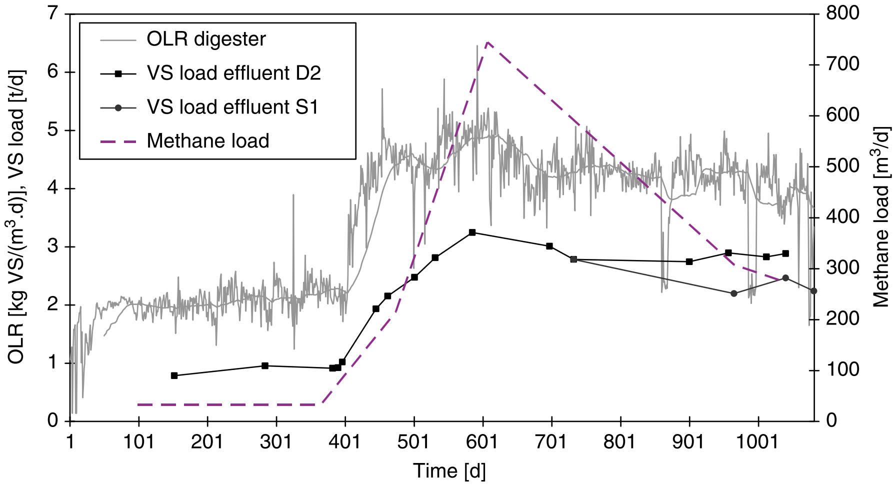

Figure 4 Dynamic of the theoretical methane production from the digestate (effluent D2) and from the digestate storage (ES)

As can be seen, the RMP in phase 2 increased to maximum values 10 times higher than in phase 1. In phase 3 the RMP decreased to a medium level again. This observation has already been made by other authors. Lehtomäki (2006) observed a residual methane potential of 2.9–7.7 m^3 CH_4/t FM at 35 °C after increasing the OLR from 2.0 to 3.0 kg VS/(m^3d). Weiland *et al.* (2004) observed a residual methane potential in operating biogas plants in Germany ($n = 11$) which was mainly lower than 10% but did reach 25% (20 °C). Such alarming levels were only observed in one-stage CSTR systems. Angelidaki *et al.* (2005) recommended, therefore, the integration of the effluent storage into the gas proof biogas collection system of the plant, and furthermore a minimum temperature in the storage of 20 °C to avoid methane emissions.

When calculating the mass flow of the digestate a theoretical daily methane production of 750 m^3 from the digestate, which corresponds to 14.4% of the total methane production

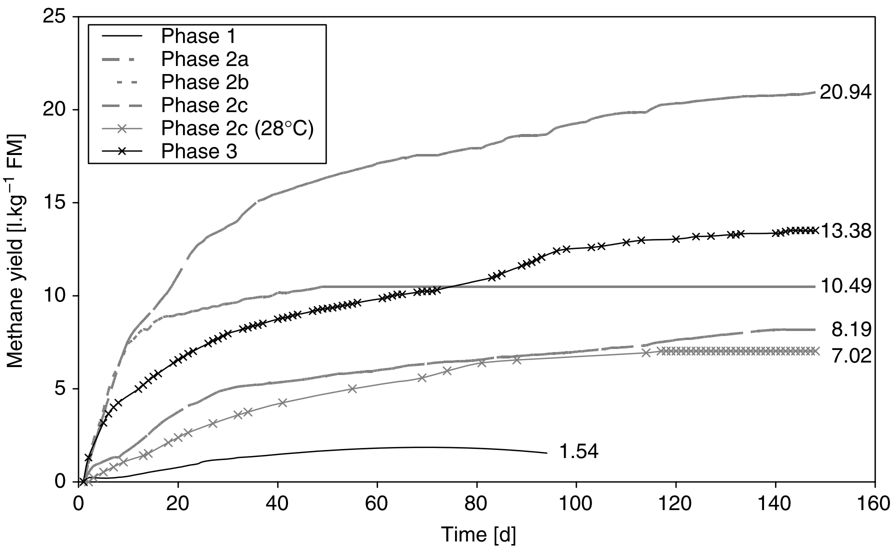

Figure 5 Methane yields (35 °C) from the digestate of D2 during the three operation phases

of the biogas plant (Figure 4) was calculated. In phase 1 this part was 1.1% and in phase 3 it was 7.9% when entering the covered storage tank and 5.5% when leaving it.

The methane production of the fermentation test at 28 °C was significantly lower than the 35 °C tests. However, the daily methane production would correspond to 251 m^3 or 4.8% of the total methane production of the plant. Above all, methane emissions out of the effluent storage should be avoided because of their global warming potential. In the design of further biogas plants this should be considered.

The agricultural usability of the effluent was affected in two ways. The ratio of TAN in TKN decreased from 71.5 to 57.2% due to the process changes. Furthermore, the higher content of VFA is a potential source of odour emissions. Other changes in the process due to the higher loading of solid substrates connected with the process enhancement, were an increase of the viscosity in the digesters and consequently a higher demand on the stirring system and the rise of the ammonium content up to 6.5 $g.L^{-1}$. This high nitrogen concentration could lead to ammonia inhibition with higher temperatures (Ahring *et al.*, 1995; Strik *et al.*, 2006). Therefore a cooling system could be necessary to prevent a temperature increase in the digesters due to the effect of self-heating (Lindorfer *et al.*, 2006).

Conclusions

The results of this study show that the additional feeding of anaerobic digesters with energy crops is a simple way to increase digester efficiency. However, some consequences in the digester performance should be expected to accompany this process enhancement. These are changes of the chemical parameters in the digester content and in the digestate, such as an increase in solids and nitrogen. The most serious development connected to the enhancement is the rising transfer of partly degraded organic material into the effluent storage. This caused a strong increase of the residual methane potential in the effluent. To avoid methane emissions due to the increasing biomass addition, the digestate storage should be covered.

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6. Comparing centralised and decentralised anaerobic digestion of stillage from a large-scale bioethanol plant to animal feed production

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Comparing centralised and decentralised anaerobic digestion of stillage from a large-scale bioethanol plant to animal feed production

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ABSTRACT

A comparison of stillage treatment options for large-scale bioethanol plants was based on the data of an existing plant producing approximately 200,000 t/yr of bioethanol and 1,400,000 t/yr of stillage. Animal feed production – the state-of-the-art technology at the plant- was compared to anaerobic digestion. The latter was simulated in two different scenarios: digestion in small-scale biogas plants in the surrounding area versus digestion in a large-scale biogas plant at the bioethanol production site. Emphasis was placed on a holistic simulation balancing chemical parameters and calculating logistic algorithms to compare the efficiency of the stillage treatment solutions. For central anaerobic digestion different digestate handling solutions were considered because of the large amount of digestate. For land application a minimum of 36,000 ha of available agricultural area would be needed and 600,000 m^3 of storage volume. Secondly membrane purification of the digestate was investigated consisting of decanter, microfiltration, and reverse osmosis. As a third option aerobic wastewater treatment of the digestate was discussed. The final outcome was an economic evaluation of the three mentioned stillage treatment options, as a guide to stillage management for operators of large-scale bioethanol plants.

Key words | anaerobic digestion, animal feed, bioethanol, digestate treatment, stillage

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INTRODUCTION

The production of ethanol is one of the major fermentation processes worldwide and is set to increase in the future owing to a surge in demand for biofuels. A contributing factor of this demand is due to political forces. The EU intends to raise renewable energy to 5.75% in the fuel sector. The Austrian government aims to raise this even to 10% by 2010. The construction of large-scale bioethanol plants will increase, therefore stillage management and stillage treatment processes will have to be explored in greater detail.

This study evaluates the stillage treatment process currently in use at an existing bioethanol plant (200,000 t/yr of bioethanol) which mainly uses corn and wheat as substrates. The standard procedure is processing stillage to dried distiller's grains and solubles (DDGS) which is used as animal feed. However, DDGS production consumes a lot of energy owing to the evaporation and drying processes. For that approximately the same amount of energy is consumed as in the entire bioethanol production process (Friedl *et al.* 2005), on top of the fact that the bioethanol production is already very energy-consuming because of the distillation process. Due to rising energy prices bioethanol plant operators will have to optimise energy consumption, to avoid a negative impact on the costs of ethanol production (Wilkie *et al.* 2000; Pfeffer *et al.* 2007). Therefore, this study investigates

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scenarios for energy co-production by simulating anaerobic digestion (AD) from the stillage produced at the bioethanol plant. Energy is produced which can replace fossil fuels at the plant. In addition, by using biogas as energy source green house gases can be reduced decisively and a better eco-balance can be achieved. This is very important since the EU is planning a certification scheme for biofuels where a minimum of green house gas reduction will be demanded.

This study focuses on AD as promising option for stillage treatment. Wilkie *et al.* (2000) have stated in a technical evaluation of various stillage treatment options that AD is economically viable and sustainable. Besides, less sludge is produced by AD than in aerobic treatment of stillage, where also significant aeration power would be required. One advantage of AD could be the possibility to use grains of lower quality for ethanol production: rye or grains contaminated with mycotoxins cannot be processed to animal feed, so if these substrates had no negative effect on ethanol production they could be used and their residues converted to biogas. If AD is concerned the efficiency of the process no longer depends on the market for DDGS but on the energy market, where the prices are expected to keep increasing. There are two main scenarios to realise AD of the produced stillage: either to build a central biogas plant at the bioethanol production site or to sell the stillage to surrounding small-scale biogas plants. Thus these two different strategies were investigated in the study comparing them to the state-of-the-art DDGS production.

METHODS

Through a modular design of the simulation process an easy comparisons was made possible (Figure 1). The model of the theoretical processes consisted of DDGS production, peripheral AD in surrounding small-scale biogas plants, or central AD. For the latter, three digestate treatment options were simulated: membrane purification, aerobic wastewater treatment, and land application as fertilizer.

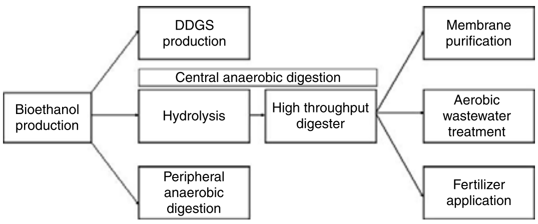

Figure 1 | Setup of the different modules in the simulation model.

Each module containing process flows was treated in a black box-approach with input and output parameters, or divided into sub-modules which were treated alike. The process flows in the model were calculated as mass and energy balances using the measured parameters. For the calculation of anaerobic digestion also data from the working group's data base were integrated into the simulation. From data provided by industry every module and sub-module could be given a financial value, so that a coherent overall picture of the simulated process options was obtained and the comparison could be made. For machinery a 10 year amortisation period was considered. Operating costs and energy costs were calculated but no costs of manpower entered into the comparison.

Characterisation of process flows

Samples were taken from the bioethanol plant to characterize the different process flows. The following parameters were obtained using *Standard Methods* (APHA 1998): Total Solids (TS), Volatile Solids (VS), and Chemical Oxygen Demand (COD). Also the nitrogen content in the process flows was considered in the simulation by Total Kjeldahl Nitrogen (TKN). The biochemical methane potential (BMP) of the stillage was evaluated through anaerobic batch fermentation tests according to the modified norm DEV S6, DIN 38 414-S6 (DEV 2005) at 35°C using as inoculum fermented sludge from a biogas plant fed by pig manure, corn whole crop silage and grass silage.

DDGS production

In the DDGS production process the stillage is separated into a solid phase (wet cake) and a liquid phase (centrifugate). The centrifugate is concentrated to syrup by vacuum evaporation. This syrup is then mixed again with the produced wet cake and dried to DDGS. For the comparison the investment costs of the machinery and the costs for carbon dioxide certificates were included. For energy costs the heat energy needed was calculated in fossil gas consumption. However, the vacuum concentration step (from about 10% TS to about 30% TS) was not considered as extra costs since internal energy from ethanol condensation is used in the process. In addition, to demonstrate the influence of rising energy prices three different scenarios were calculated: present energy costs, a 50% rise in energy costs and a 100% rise. For the DDGS price two scenarios were calculated. First a price of 200 €/t which was about the price assumed in Friedl *et al.* (2005), secondly, a lower price of 100 €/t. The reason for this was that bioethanol production is increasing worldwide and with an increase of DDGS on the market the price could be lower.

Central AD

Because of the large quantities of stillage (1,400,000 t/yr) an innovative scheme for high throughput was presented and calculated in this study. Therefore a pre-treatment step with microbiological hydrolysis was considered where part of the solid substances is liquefied. The remaining solids are separated by a decanter and converted to compost. For composting additional costs of 40 €/t were calculated. The liquid hydrolysed phase is fed into high throughput digesters. The produced biogas is converted to energy by combined heat and power units. For the calculation of the revenue the actual energy prices paid by the bioethanol plant were considered. For the digestate treatment three different scenarios were calculated, as described here.

Fertilizer application

In this module the surrounding agricultural areas at the production site were considered with an application limit for nitrogen of 210 kg N/ha (according to national regulations). Since digestate cannot be applied all year over a storage capacity of half a year had to be considered, calculating a 10 year amortisation period for storage containers. Digestate transportation and application was calculated assuming 20 m^3 application carts. The benefit of digestate fertilizing was calculated from nitrogen (N), phosphorus (P) and potassium (K) content of the digestate.

Membrane purification

Here the setup and the costs were calculated according to the membrane treatment process presented in Klink *et al.* (2007). This module therefore consists of a decanting step, and consecutive purification by ultrafiltration followed by three steps of reverse osmosis.

Aerobic wastewater treatment

Lacking experimental data of an existing biogas plant joined with an aerobic wastewater treatment plant, this module was estimated from the expenses of a standard large-scale aerobic wastewater treatment plant with an amortisation period of 15 years. In addition, the costs of an additional carbon source were calculated since it is needed to achieve entire denitrification. The reason for this is that most of the organic matter is metabolised in AD, whereas a high nitrogen concentration remains in the digestate. Because bioethanol is present at the plant in great quantities, it was taken as external carbon source in the simulation. In addition, it poses a good carbon source and it is easy to calculate its costs. Nevertheless in practise alternative cheap carbon sources have to be identified and used. About 3–5 kg CSB of carbon are needed for the denitrification of 1 kg nitrate (Gerardi 2002). According to these values two different scenarios of the addition of ethanol were calculated: First the addition of ethanol-CSB to TKN in the relation 1:1, second in the relation 2:1. The ethanol price was taken as 640 €/m^3.

Peripheral AD

In this module the stillage is digested in small-scale (500 kW$_{el}$) biogas plants in the surroundings. Every biogas plant was calculated as its own profit centre, to guarantee a realistic stillage price that the biogas plant operators are able to pay. In order to raise the stillage price, transportation costs and logistics were considered to be the responsibility of the bioethanol plant operator. This option was evaluated by determining revenue of stillage sale and costs of stillage transportation.

RESULTS AND DISCUSSION

The methane potential and the characteristics of the corn and wheat stillage produced at the plant are presented in Table 1. The nitrogen (TKN) level in wheat stillage was

Table 1 | Characteristics of corn and wheat stillage

	pH	COD [g/kg]	TS [%]	VS [%]	TKN [g/kg]	BMP [m^3/t VS]
Corn stillage	4.7	159	9.7	9.1	4.5	611
Wheat stillage	5.5	147	10.2	9.3	6.9	579

quite high with 6.9 g/kg which could have a negative effect on process stability when carrying out continuous fermentation. For a more realistic approach to estimate the methane production in the continuous process only 75% of the measured maximum methane potential was assumed in the following comparison.

DDGS production

At the bioethanol plant about 4,022 t/d of stillage are produced and processed to about 415 t/d of DDGS (mounting to over 145,000 t/yr). The calculated process flows are presented in Figure 2 which were used for calculating the mass and energy balance.

The results of the economic evaluation of the DDGS production are given in Table 2. For the scenario where a DDGS price of 200 €/t is assumed the margin is 22.4 million € (Mil. €)/yr and for the second (100 €/t DDGS) 7.9 Mil. €/yr. This demonstrates the great influence of the DDGS price on the evaluation. Especially in the 100 €/t DDGS scenario you see the great difference concerning rising energy costs: Considering present energy costs the margin is 7.9 Mil. €/yr, if energy costs rise 100% the margin drops to less than a half (3.0 Mil. €/yr).

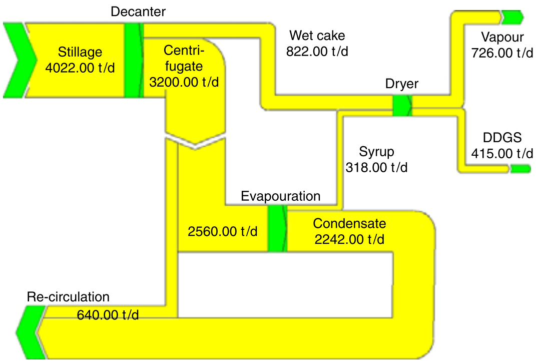

Figure 2 | Process flows in DDGS production.

Central AD

The large-scale biogas plant would mount to about 21 MW_{el} and 25 MW_{therm}. In Figure 3 the simulated process flows are shown, using membrane technology as digestate treatment. The expenses for AD (without calculating digestate treatment) are 7.2 Mil. €/yr, about one third (2.3 Mil. €/yr) are the costs for composting the not hydrolysed solids. If this fraction could also be used as fertilizer, the expenses for AD would be quite lower. The total revenue for electricity and heat is 19.8 Mil. €/yr.

Digestate treatment

In Table 3 the expenses for the digestate treatment scenarios are presented. For land application as fertilizer 1,200,000 t/yr of digestate have to be applied on a minimum of 36,000 ha of agricultural area and 600,000 m^3 of storage volume is needed. Storage costs are 3.1 Mil. €/yr and transportation and application costs are 6.4 Mil. €/yr, total expenses are 9.5 Mil. €/yr. The fertilizer value due to the N, P and K content would mount to 5.5 Mil. €/yr, leaving total expenses of 4.1 Mil. €/yr. Since it is not credible that agriculture would pay the same price for digestate as for fertilizer concentrate, realistic expenses would lie somewhere between 4.1 and 9.5 Mil. €/yr. In addition the local nutrient demand or surplus has to be considered.

The expenses of membrane purification (5.1 €/t digestate; Klink *et al.* 2007) mounts to 6.3 Mil. €/yr. In the simulation the RO-concentrate (739 t/d, see Figure 3) was assumed as fertilizer without cost or profit, although it has quite a high salt-concentration, so the fertilizing qualities could be questionable.

In aerobic wastewater treatment the expenses turn out to be 5.9 Mil. €/yr (calculating the relation of ethanol COD to TKN to be 1:1) or 9.4 Mil. €/yr (considering a relation of 2:1).

Table 2 | Expenses, revenue and margin for DDGS production in dependence on DDGS-price and energy costs

	DDGS price of 100 €/t			DDGS price of 200 €/t		
	Expenses Mil. €/yr	Revenue Mil. €/yr	Margin Mil. €/yr	Expenses Mil. €/yr	Revenue Mil. €/yr	Margin Mil. €/yr
Today's energy prices	6.6	14.5	7.9	6.6	29.0	22.4
50%-rise	9.0	14.5	5.5	9.0	29.0	20.0
100%-rise	11.5	14.5	3.0	11.5	29.0	17.5

Evaluation of central AD and digestate treatment

For the final comparison membrane purification (with expenses of 6.3 Mil. €/yr) was selected as standard process for central AD. This means a margin of 6.2 Mil. €/yr for central AD. Membrane purification is preferable to fertilizer application on the one hand because more than 950,000 t of water per year are recycled into the process by membrane purification, on the other hand yearly expenses for land application vary a lot depending on the realistic fertilizer value of the digestate. Finally, fertilizer application of 1,200,000 t/yr is a great logistic challenge, the logistics of membrane purification are more easy to handle for a bioethanol plant operator. Membrane purification of digestate is also preferable to aerobic treatment, because the rather high cost of an external carbon source can be avoided. The carbon (organic matter) of stillage is removed to a great part by AD, so it makes little sense to add again expensive C-source to achieve a stable aerobic treatment. The cheaper one of the two aerobic treatment scenarios produces expenses of 5.9 Mil. €/yr which is comparable to the costs of membrane purification (6.3 Mil. €/yr). Since the latter is considered to be more stable and costs are about the same or lower than in aerobic treatment, membrane purification was finally chosen as standard digestate treatment process for the model. Also combinations of aerobic treatment and membrane purification could be a viable solution.

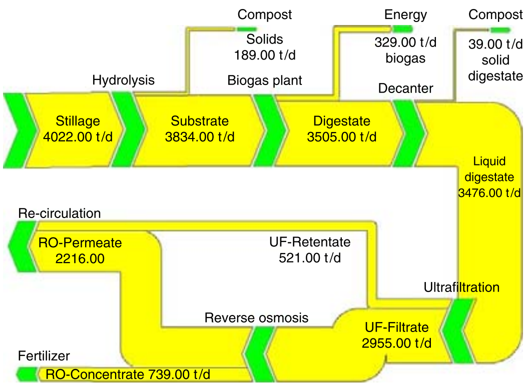

Figure 3 | Process flows of simulated central AD with membrane purification of digestate.

Peripheral AD

This scenario means that the produced stillage would be transported to about 38 smaller-sized nearby biogas plants (with 500 kW_{el} and $\eta_{el} = 0.35$), at a presumed average distance of 40 km. In the province of Lower Austria where the bioethanol production site is located there exist 100 biogas plants with an installed total capacity of 33.95 MW_{el} (E-Control GmbH 2007). So at least 56% of the energy produced by AD in this province would be provided by the stillage of the bioethanol plant. A realistic price of 1.2 €/t stillage was assumed which results in a revenue of 1.6 Mil. €/yr for the operator. To achieve this price transportation costs (of about 8.1 Mil. €/yr) would have to be covered by the bioethanol plant operator. Thus the total expenses of

Table 3 | Expenses of the different scenarios for digestate treatment

	Expenses [Mil. €/yr]
Fertilizer application (including theoretical fertilizer value)	4.1
Fertilizer application (w/o fertilizer value)	9.5
Membrane purification	6.3
Aerobic wastewater treatment (ethanol-COD:TKN = 1:1)	5.9
Aerobic wastewater treatment (ethanol-COD:TKN = 2:1)	9.4

peripheral AD are 6.5 Mil. €/yr. Since the other two stillage treatment options produce earnings instead of costs, peripheral AD should be disregarded as an option for large-scale bioethanol plants. In small-scale or middle-size bioethanol plants this option can still be a good choice, since by peripheral AD no digestate management is needed which is one of the key issues in central AD and quite cost intensive.

Outcome of the comparsion

The results of the comparison (Table 4) show that under the described conditions (today's energy prices) it is favourable for operators of large-scale bioethanol plants to produce DDGS. However, this is only the case when DDGS can be sold above 100 €/t. If the DDGS price is significantly below this level AD becomes competitive. At a DDGS price of 100 €/t the yearly margin will be about 7.9 Mil. €/yr for DDGS production, whereas central AD will earn 6.3 Mil. €/yr. Additionally, if energy prices rise by 50% DDGS production will only produce 4.5 Mil. €/yr. So the tendency of DDGS prices and energy prices should influence a plant operator's decision to introduce AD. Rising energy prices always favours AD, since the energy consuming distillation process can be fed with the energy produced and no expensive external energy has to be purchased. If energy prices rise 100% the margin for DDGS production becomes decisively lower than for AD: At a DDGS price of 100€/t the margin will be 3.0 Mil. €/yr and at 200 €/t it will be 17.5 Mil. €/yr. Since AD produces energy its margin is higher than both DDGS production scenarios:

Table 4 | Comparison of expenses, revenue and margin for DDGS production, central AD (with membrane purification) and peripheral AD (including rise of energy prices)

	Margin [Mil. €/yr]
Today's energy prices	
DDGS production (100 €/t DDGS)	7.9
DDGS production (200 €/t DDGS)	22.4
Central AD	6.2
Peripheral AD	−6.5
100% rise in energy prices	
DDGS production (100 €/t DDGS)	3.0
DDGS production (200 €/t DDGS)	17.5
Central AD	25.1

The earnings will be 25.1 Mil. €/yr. This shows that even at a higher DDGS price AD stays the preferable option when energy prices rise decisively.

This study also reveals that at large-scale bioethanol plants central AD is preferable to peripheral AD. The latter produces a minus of 6.5 Mil. €/yr, considering that only a price of 1.2 €/t (stillage) can be paid by small-scale biogas plants to assure their positive balances and that the bioethanol plant operator must cover the transportation costs. Central AD, however, earns 6.2 Mil. €/yr at today's energy prices.

CONCLUSIONS

At today's energy prices DDGS production is preferable to AD. Only if the DDGS-price drops below 100 €/t AD becomes competitive. If energy prices rise decisively biogas production will be both an economically and ecologically better option than animal feed production, even if DDGS prices are 200 €/t. The main problem for central AD at a large-scale bioethanol plant is the digestate treatment. As this study shows over 50% of the costs for AD will be due to digestate treatment. Here the treatment by separation processes containing membrane purification seems to be one of the preferable solutions. It is a stable process and the costs are comparable or cheaper than fertilizer application or aerobic wastewater treatment. The results show clearly that the AD in surrounding small-scale biogas plants is no economically viable solution. So if AD is concerned central AD at the plant location is the option to chose. Therefore further research should focus on digestate management. Still data from large-scale anaerobic digesters with implemented membrane purification are lacking. So before membrane technology becomes standard technology at biogas plants, however, additional research on this topic should be carried out.

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7. Full scale application of anaerobic digestion of slaughterhouse wastes – long term experiences, problems and resulting strategies.

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Full scale application of anaerobic digestion of slaughterhouse wastes in biogas plants – long term experiences, problems and resulting strategies.

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Abstract
In this contribution a full scale biogas-plant run with blood, colon, gut and fat scrubber content is described in a course of a four year-period of operation. The methane produced from slaughterhouse wastes covered almost half of the adjacent abattoirs` energy demand. The semi-continuous operation of the biogas plant results in a characteristic weekly pattern of methane concentration and productivity.
High values of Ammonia result through anaerobic digestion from the high protein content of the substrate and represent approx. 82 to 86% of total nitrogen. High ammonia-levels cause high concentrations of acid intermediate-metabolites such as volatile fatty acids. The concentrations of (unionized) free ammonia (NH_3), believed to be the toxic part of total ammonia, are calculated based on simple dissociation-constants and by equilibrium-simulation taking major acid components into account. Based on simple dissociation-constants unrealistic high values of free ammonia are calculated. Taking ion-activities and acid components as counter-ions to the ammonium-ion into account, the real niveau of free ammonia is in the range of 100-300 mg/l which corresponds perfect to the first values published in the context of ammonia-toxicity. This results suggest that high concentrations of volatile fatty acids are required to keep values of free Ammonia low and therefore may not be used as process stability indicators.

Keywords: anaerobic digestion, slaughterhouse wastes, ammonia, nitrogen, toxicity, volatile fatty acids, biogas

Introduction
Rendering of animal proteins has been an accepted pathway for treatment of slaughterhouse wastes for a long time. Due to the occurence of BSE in Europe, the European Commission banned rendered animal protein as a feed for livestock in 2000 (European Commission, 2000). This increased gate fees for the treatment of slaughterhouse wastes on the one hand and resulted in a new regulation from the European Commission and the European Parliament laying down health rules regarding animal by-products (ABPs) not intended for human consumption (European Commission, 2002).
Only such parts of animals devoid of presumable BSE or TSE-infectious tissues may be processed in biogas plants. Experiments have shown that the degradation of infectious

proteins (“Prions”, Prusiner, 1982) may be insufficient under anaerobic conditions (Kirchmayr *et al.*, 2006)

Annually more than 16 million tons of animal material not intended for human consumption, the bulk of which derive from healthy animals, are produced in the EU (European Commission, 2010). In Austria 582,000 tons of ABPs per year (including catering waste) need to be treated. From there 347,000 t is slaughterhouse waste (derived directly from slaughtering and meat processing industry) which equals around 40 kg per capita and year (Lebensministerium, 2010).

Regulation (EC) No 1774/2002 and Regulation (EC) No 1069/2009 - replacing the latter, explicitly comprise treatment of slaughterhouse waste (ABP’s) in biogas plants define parameters for the operation of such biogas plants. Treatment parameters principally cover general hygienic rules and comprise an obligatory pasteurisation or hygienisation unit (treatment at minimum 70°C for 60 minutes with maximum particle size of 12 mm or a corresponding validated process).

Based on this clear European legislation, several abattoirs deliver ABP’s to biogas plants which treat slaughterhouse waste in codigestion with other wastes. Only sparse information is available on slaughterhouses having constructed biogas plants to treat their own wastes and to gain energy for their facilities.

Rudolf Grossfurtner GmbH, Austria’s biggest pig abattoir runs a biogas plant operated solely on ABP’s of pigs such as blood, colon, fat scrubber content (after dissolved air flotation). Rumen content is obtained from the nearby bovine slaughtering facility. In the actual plant setup approx. 4.7 MWh/d of electricity and 7 MWh/d of heat can be produced. The electricity is fed into the national power grid and the heat is delivered to the slaughtering facility. The specific heat-load-profile of the slaughterhouse showed that only during 35% of the time the heat produced by the combined heat and power plant (CHP, run on the biogas produced in the biogas plant) could directly be consumed in the slaughterhouse. Therefore a hot water storage tank serves as a buffer to match the steady heat production with the heat demand of the slaughterhouse. Thus nearly all of the heat produced in the biogas plant is consumed in the slaughterhouse, covering 45 % of its thermal energy demand (Kirchmayr *et al*., 2009).

Operating a biogas plant with ABP’s requires sensitivity, not only in the field of hygiene and toxicity of ammonia-nitrogen, but also regarding odour emissions (Kirchmayr *et al.*, 2007). The manipulation hall, the buffer tank and eventually open storage lagoons have to be connected to a powerful exhaust air treatment. Alternatively the latter two tanks are connected to the gas collection system due to the emission of hydrophobic substances. Even the digestate used as agricultural fertilizer may cause nuisances, which have to be overcome by low-emission spreading.

In this pig slaughtering facility around 5 l of blood, 6 l of hind gut (and its content) and 10 l of fat scrubber content (after dissolved air flotation) per slaughtered animal are obtained and treated in the biogas plant. These figures also include small amounts of process water and are therefore higher compared to those reported by Freudenreich and Bach, 1993.

Pilot scale applications and experiences of anaerobic digestion of slaughterhouse wastes (Edstöm *et al.*, 2006, Hejnfeldt and Angelidaki, 2009, Lopez *et al.,* 2006, Siegrist *et al.*, 2006, Wu *et al.,* 2009,) have already been reported, but no full-scale application has been described so far.

High concentrations of free ammonia cause microbial inhibition. Inhibition of the biogas process is related to total ammonia concentration (Kayhanian, 1999; Sprott and Patel, 1986; Wiegant and Zeeman, 1986) and free or unionized Ammonia (NH_3, UAN – unionized ammonia nitrogen), which is considered to be the toxic form (McCarty and McKinney 1961). Between the different strains of micro organisms involved in anaerobic digestion, the methanogens are the least tolerant and therefore most likely to be inhibited (Koster, 1986, Koster 1988, Angelidaki and Ahring, 1993, Braun et al, 2010, Schnurer and Nordberg., 2008).

Practical experiences with full-scale bio-waste and slaughterhouse waste digesters show that ammonium concentrations exceeding 5 g/L NH_4^+-N still allow stable operation (Kirchmayr et al. 2007; Resch et al. 2006). In the biogas plant described here the monitored concentration of ammonium nitrogen (NH_4^+-N) in the fermenters ranges (depending on the substrate throughput and composition) from 5.3 to 8.5 g/L. Such high levels cause microbial inhibition, drops in the methane potential (Angelidaki and Ahring, 1993, Hansen et. al. 1998) and high concentrations of volatile fatty acids. Low concentrations of volatile fatty acids (VFA) are considered as a common indicator of process stability (Ahring *et. al., 1995*). Higher values of VFA have to be evaluated in combination to present ammonia concentrations. In this contribution it is shown that high concentrations of ammonia are causing high concentrations of volatile fatty acids which act as acid counter-ion to the ammonium-ion and therefore are necessary to keep values of free ammonia (NH_3) low. Free Ammonia is a function of Ammonium (NH_4^+) concentration, pH, CO_2, the concentration of volatile fatty acids and the digester temperature. Thus high concentrations of VFA are not an indicator for severe process instabilities in the context with high ammonia-concentrations.

Material and Methods

The measurement of volatile fatty acids was performed with HPLC (Agilent 1100, with RI- and WL-detection) after precipitation according to the method of Carrez to minimize protein-concentration. Total Nitrogen (TKN) and Ammonia-Nitrogen (Kjeldatherm, Gerhard GmbH), pH (340i, WTW), total solids (TS), volatile solids (VS) and chemical oxygen demand (COD) were determined using standard methods (APHA, 1998)

Anaerobic Batch-Test analysis for determination of maximum methane yield was carried out based on the regulation VDI 4630.

Simulation calculations to obtain values of unionized ammonia (considering the concentrations of CO_2 and volatile fatty acids) were carried out using ASPEN PLUS.

Results and Discussion

1. Description and operation of Biogas plant

The plant design corresponds to the classical concept of an agricultural biogas plant consisting of two main fermenters of 600 m³ (Fermenter 1) resp. 1,000 m³ (Fermenter 2), a secondary fermentation tank (without heating, 1,000 m³; Fermenter 3) and a storage tank (3,200 m³). The biogas plant is operated with selected fractions of the pig slaughtering process such as pig blood, minced hind gut including content and fat scrubber content (after dissolved air flotation, DAF). The latter substrate characteristics changed during the monitoring period due to optimization of the DAF (Table 2). According to the European Regulation (EC) No. 1774/2002 and Reg. (EC) No 1069/2009, the fat scrubber content has a maximum particle size of 6 mm and the other substrate fractions are crushed to a maximum particle size of 12 mm. The substrate is pumped from the slaughterhouse to a buffer tank and subsequently pasteurised (70°C/60 min). Before feeding, the substrate is cooled to 55°C

to minimise potential damage of bacteria in the biogas fermenter. On average, approx. 20 m³ of slaughterhouse wastes per day are fed to the two digesters in parallel (Table 1).
Rumen content from the neighbouring bovine abattoir is delivered to the biogas plant and by means of a feeding screw brought into the smaller digester. Both digesters are operated at mesophilic temperatures (35°C). Depending on the substrate composition ammonia-nitrogen levels vary between 5.3 and 6.2 g/L in fermenter 1 and between 6.0 and 7.4 g/L in fermenter 3 respectively, corresponding to a TKN concentration between 7.1 and 8.5 g/L (Table 5 and Figure 1).

Table 1: Average substrate mass streams. Monitoring period 4 years (1460 days). Average values are arithmetic means within days of addition of the specific substrate.

	Flotation fat (disloved air flotation)	Pig Blood	Hind gut of pig (incl. content)	Bovine Rumen Content	Anaerobic Sludge (Sewage)
Days of feeding [d]	1003	816	1003	488	123
Loading rates					
Average [m³/d]	16.85	8.94	10.60	6.78	13.93
Std deviation [%]	33.2	15.6	17.2	25.4	33.5

Loading rates may be calculated based on the real-case-scenario of feeding 5 days per week or as weekly mean values. Table 3 shows that the real daily organic loading rate (5 days/week feeding) of the biogas plant is 5.07 kg/m³.d VS (10.8 kg/m³.d COD) in fermenter 1 and 2.95 kg/m³.d VS (6.28 kg/m³.d COD) in fermenter 2 respectively.
Blood (as the main source of nitrogen) is added in function of the ammonia concentration in the digesters. High levels of ammonia lead to high concentrations of volatile fatty acids, indicating a poor microbiological activity and leading to foaming problems. If fermentation problems occur, anaerobic sludge (from a near-by sewage sludge digester) is used to dilute the fermenter content and to reinoculate the process (Table 1). During disturbances addition of blood is as substrate is seized and is retained in a rendering plant until the stability of the digestion process has been reestablished.

Table 2: Chemical characterisation and anaerobic batch-test degradation analysis of substrates

		Blood	Hind gut &Content	Fat scrubber 2006	Fat scrubber 2010	Rumen content
COD	[g/kg]	389.0	575.0	99.1	157.1	197.7
TS	[%]	20.75	23.04	8.27	6.12	18.02
VS	[%]	19.72	21.86	7.17	5.43	16.66
TKN	[g/kg]	22.78	11.12	4.36	1.27	4.59
$Y(CH_4)$ FM	[Nm³/t]	91.4	200.4	32.2		58.2
$Y(CH_4)$ COD	[Nm³/t]	234.4	265.0	325.1		294.6
$Y(CH_4)$ VS	[Nm³/t]	463.7	458.3	449.3		349.6

Legend: $Y(CH_4)$ ... Methane yield or biological methane potential

Table 3: Organic loading rates of the whole plant and fermenter 1 and 2 in particular calculated based on a feeding of 5 days a week (operation of the plan) and as weekly averages.

feeding days/week			fermenter 1 [kg/m³.d]	fermenter 2 [kg/m³.d]	total plant [kg/m³.d]
5	operation	COD	10.77	6.28	4.82
5	operation	VS	5.06	2.95	2.26
7	Weekly average	COD	7.36	4.30	3.29
7	Weekly average	VS	3.48	2.03	1.56

2. Chemical fermentation parameters

The start-up period in 2002 was accompanied by several serious fermentation failures resulting in bad degradation and foaming. Subsequent problems due to odour emission required a chemical monitoring process. Volatile fatty acids (VFA, C_2-C_5), as a common indicator of process stability (Ahring *et. al., 1995*) the Ammonia-Nitrogen, the Total Nitrogen (TKN), total solids (TS) and volatile solids (VS) were measured in parallel. In the range of values present here, measurements of volatile fatty acids with laboratory standard methods (HPLC) and quick photometric tests or titration methods showed quite a good correlation and therefore could be used as monitoring tools directly at the biogas plant (Porak, 2006).

Figure 1 shows the concentrations of volatile fatty acids (VFA) and the corresponding concentration of ammonia-nitrogen. Ammonia-Nitrogen concentrations measured in fermenter 1 are slightly lower than those in fermenter 2 and 3 respectively. There is a very strong correlation between ammonia-nitrogen and TKN in each fermenter (Standard deviation 3%). The percentage of Ammonia-Nitrogen of TKN increases along the fermenter cascade from (median) 81.6% in fermenter 1 to 84.9% in fermenter 2 up to 86.1% in fermenter 3 and represents the different mineralization rates of proteins in the specific fermenter. The difference between fermenter 1 and fermenter 2 is due to the fact that the effluent of fermenter 1 is pumped to fermenter 2, which is additionally fed with substrate.

The concentration of volatile fatty acids present in this biogas plant exceeds the values recommended for the operation of biogas plants (Lindorfer *et. al.*, 2008, Weiland, 2004). Most of Austrian biogas plants operated with energy crops show VFA concentrations less than 2 g/l. Figure 1 shows that VFA concentrations exceed the 2 g/l limit in most of Austrian biogas plants operated with energy crops (Gabauer *et. al.*, 2008). Median values of VFA in fermenter 1 and fermenter 2 are similar with 12.6 g/l and 13.9 g/l respectively (Table 4), reaching up to maximum levels of 24.5 g/l in fermenter 1 (Figure 1). The median of VFA in fermenter 3 is characteristically lower at 8.3 g/l.

Table 4: Mean values of volatile fatty acids in the specific fermenter.

	Fermenter 1			Fermenter 2			Fermenter 3		
	Acetic Acid [mg/l]	Propion. Acid [mg/l]	VFA total [mg/l]	Acetic Acid [mg/l]	Propion. Acid [mg/l]	VFA total [mg/l]	Acetic Acid [mg/l]	Propion. Acid [mg/l]	VFA total [mg/l]
Median	5,897	4,935	13,470	5,914	5,299	13,880	3,279	4,003	8,259
Max	11,817	6,487	24,460	9,521	6,668	21,001	5,673	6,063	12,966
Min	1,546	1,754	5,733	2,708	1,860	6,727	532	255	959

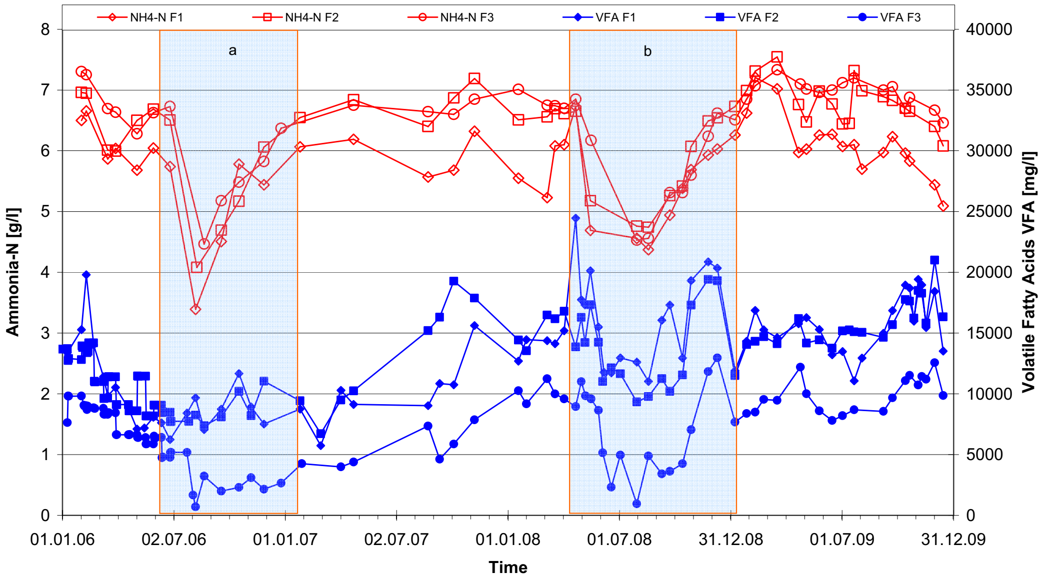
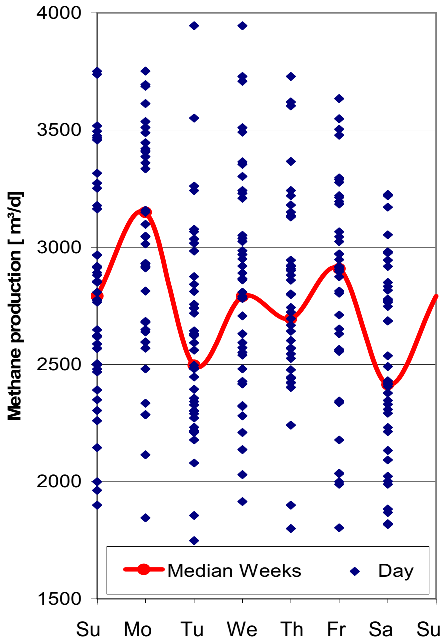


Figure 1: Concentration of Volatile Fatty Acids (C_2-C_5) and Ammonia-Nitrogen of all 3 Fermenters during a monitoring period of 4 years

3. Discontinuous operation

Figure 2 shows values of gas quality and methane productivity related to the days of the week. As this biogas plant is run semi-continuously (continuous substrate feed from Monday noon to and Saturday noon and no substrate feed on weekends) there is a characteristic change in gas quality during the week. On Friday the CO_2-content of the biogas is app. 2.5% higher compared to Monday. Due to the semi-continuous substrate addition the methane productivity shows a maximum on Monday and a minimum on Tuesday and Saturday. The reduction in methane productivity from Monday to Thursday shows a remarkable difference of 20.7%.



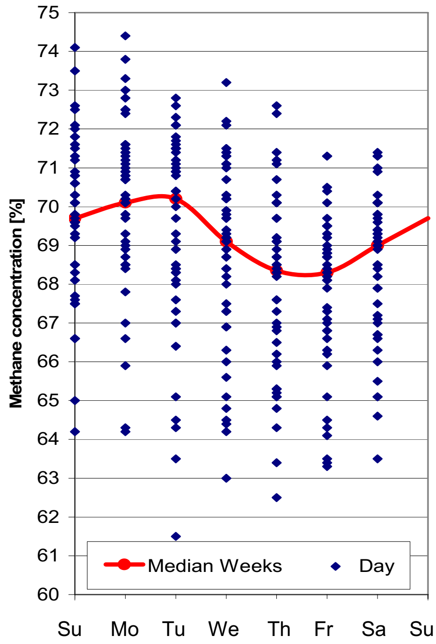


Figure 2: Change of methane productivity (left) and gas quality (right) during a week (Monitoring time: 1 year)

4. Correlation between chemical fermentation parameters and methane recovery

Based on batch-test analysis of the substrate (Table 2) and daily feed the maximum methane production of the biogas plant is calculated. Figure 3 shows the maximum methane yield as weekly and monthly averages and as median values of the last 70 days (with good fit to the curve of monthly averages). The factual methane produced was measured by gas measurement (quantitiy and quality) and calculated based on the daily electricity production of the engines.

Based on the daily substrate-feed and the substrate analysis (Table 2) the maximum methane yield of the biogas plant is calculated and shown in Figure 3 as weekly and monthly averages. Measured methane production (gas measurement and electricity produced by the gas engine) is shown as monthly averages. The calculated recovery rate in percent produced of maximum methane production is also shown as monthly averages.

Due to reduced blood addition and high water intake there are two periods with lowered NH_4^+-N values (marked as periods a and b in Figure 1). Based on these deviant Ammonia (and TKN)-concentrations a probable correlation between milieu conditions and gas production may be calculated. Comparing maximum methane yield and factual methane produced (shown in Figure 3 as recovery percentage) over the monitoring period of 4 years a fair correlation can be identified between recovery percentage and milieu conditions such as TKN, NH_4^+-N, free ammonia NH_3-N or volatile fatty acids. Although, regression analysis shows a tendency of decreasing daily gas yield and increasing volatile fatty acids on increasing Nitrogen (TKN; NH_3-N or NH_4^+-N) concentrations.

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Only values in Fermenter 3 show a reasonable correlation between the concentration of NH_4^+-N and volatile fatty acids. Linear regression analysis shows a coefficient of determination for Fermenter 3 of $r^2=0,4712$ (F2: $r^2=0,057$; F1: $r^2=0,1608$).
The analysis of correlation between Nitrogen content (TKN, NH_4^+-N or free NH_3-N) and Gas or Methane production or gas quality shows values of the coefficient of determination below 0.1.

The influence of ammonia-nitrogen (NH_4^+) or free ammonia (NH_3) has been described under batch conditions using increasing concentrations of volatile fatty acids as an indicator for process instabilities (Angelidaki and Ahring, 1993, Edström *et al.*, 2003). The concentration of free ammonia (shown in Figure 3) has been calculated based on two methods. Based on chemical dissociation constants, pH-values and ammonia-concentrations quite high values of free ammonia concentrations are the result. These values will not represent the real concentrations present in the liquid because they neglect the acid components and gas bubbles present in the liquid. Under continuous fermentation conditions carbamates and other acid components such as volatile fatty acids will reduce the concentration of free ammonia due to their counterion-character to NH_4^+. In a steady state fermentation situation there will be an equilibrium of volatile components such as volatile fatty acids, ammonia and CO_2. Using a process-simulation software (ASPEN PLUS) the equilibrium concentrations of free ammonia were calculated. These values consider the equilibrium between liquid and gaseous phase (based on the law of Henry) and acid components (all forms of CO_2, Acetic- and Propionic Acid).
Figure 3 and Table 5 show that the actual concentrations of free ammonia (taking acid components into account) are in a very constant range between 100 and 200 mg/l. In comparison the simple calculation results in values ranging from 500 to 2000 mg/l. Latter values would by far exceed reported limits of NH_3-toxicity. Values resulting of the simulation are close to those inhibition values described for unadapted cultures (Braun et al., 1981, de Baere et al., 1984).
As described before there seems not to be a remarkable correlation between measured chemical fermentation parameters such as VFA, ammonia, etc. and methane recovery rate. The periodic variability of the methane recovery rate (see Figure 3) could depend on other operation conditions such as mixing, chemicals such as cleaning agents, etc.
In fact this result suggests that the high concentrations of volatile fatty acids observed are not an indicator for major process instability but are necessary to keep the concentrations of free ammonia at a very low level. The process of anaerobic digestion of high nitrogen containing substrates may described as a self-establishing equilibrium between the alkaline ammonia-ions (NH_4^+) and the acid carboxyl-group of VFA. Both values have to be taken into consideration for a proper bench-mark of these chemical fermentation parameters.

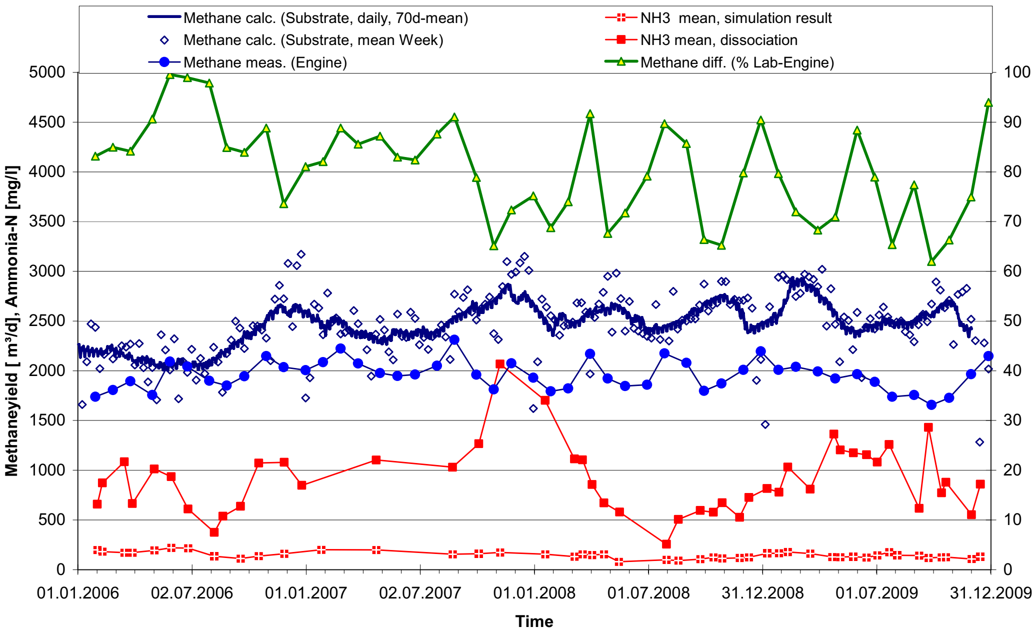

Figure 3: Methane production ("Methane calc"based on substrate addition and "Methane meas" based on the gas measurement and engine) and recovery rate in the biogas plant in correlation to free ammonia concentrations. The concentration of free Ammonia-Nitrogen (NH_3-N) is shown as mean of all 3 Fermenter based on the simple dissociation equation (high values) and also as the result of the ASPEN-simulation taking CO_2 and VFA-concentrations into account.

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Table 5: Overview of the Fermentation Data (monitoring period of 4 years). NH_3 calc. values represent values based on simple dissociation, NH_3 simul. values represent values of free ammonia after equilibrium simulation (taking acid components into account).

			Median	Std.dev. [%]
Fermenter 1	TKN	[mg/l]	7340	10,4
	NH_4-N	[mg/l]	5946	12,0
	NH_3 calc.	[mg/l]	626	52,0
	NH_3 simul.	[mg/l]	143	30,5
	pH	[ ]	8,06	3,9
	TS	[%]	4,34	26,1
	VS	[%]	3,41	29,8
Fermenter 2	TKN	[mg/l]	7800	8,9
	NH_4-N	[mg/l]	6540	11,7
	NH_3 calc.	[mg/l]	949	44,6
	NH_3 simul.	[mg/l]	174	23,4
	pH	[ ]	8,16	2,9
	TS	[%]	3,52	23,2
	VS	[%]	2,71	23,6
Fermenter 3	TKN	[mg/l]	7829	9,7
	NH_4-N	[mg/l]	6690	11,4
	NH_3 calc.	[mg/l]	856	36,7
	NH_3 simul.	[mg/l]	118	19,0
	pH	[ ]	8,12	1,8
	TS	[%]	2,99	22,8
	VS	[%]	2,08	29,7

5. Further Laboratory Experiments

The influence of high nitrogen concentrations on the micro-organisms involved in the anaerobic degradation cascade has been described for a long time (Kroeker et. al, 1979 and van Velsen, 1979).

To overcome nitrogen influence two strategies were assessed in laboratory-scale: widening of the tight C:N ratio by adding a further carbon-source on the one hand or reducing ammonia-nitrogen by simple stripping on the other Providing additional carbon sources to the sludge of fermenter 3 from previously described biogas plant by means of glycerine, starch and/or fat in continuous (in a 2 L lab-scale fermenter) and discontinuous (batch, 2 L) experiments did not increase biogas yield nor reduce volatile fatty acids (Resch et al., 2007). By means of a laboratory-scale steam-stripping column the ammonia-nitrogen concentration of the sludge in fermenter 2 was reduced stepwise to 4 g/l. Using this as a substrate to feed a lab-scale fermenter, varying the organic loading rate and hydraulic retention time (using sludge of fermenter 2 as inoculum) a substantial reduction of volatile fatty acids (90%), enhanced degradation of COD (46 % or 100 % of degradable fraction, based on batch-test assays) and a significant increase in the methane yield up to 41% (at TKN-values of 4 g/l) compared to the reference could be observed (Resch et al., 2010, in preparation).

Using the nutrient- and trace-element concentration of an anaerobic biogas-community (Scherer *et al.*, 1983) slaughterhouse wastes show a deficiency especially in cobalt, nickel and boron. By addition of atrace element solution in 20 fold excess to the sludge of fermenter 2 of the previous described biogas plant used as a substrate for a continuously operated lab scale fermentation (6L) of sludge from fermenter 3 the concentration of volatile fatty acids was reduced below 2 g/L within 2 months. The reference scenario without trace element addition showed levels of VFA comparable to the full scale fermenter.

Conclusions

The handling of a biogas plant operated with slaughterhouse wastes (ABP's) requires high sensitivity with regard to hygiene, odour emissions and the influence of ammonia to the activity of micro-organisms. This example shows that the operation of a biogas plant solely with slaughterhouse wastes is possible and may cover half of the energy demand of the abattoir.

The operation of this biogas plant shows that anaerobic digestion of high protein wastes under sub-optimal (inhibited steady state), but stable conditions is feasible. The micro-organisms will adapt to ammonia-nitrogen (NH_4^+-N) concentrations up to approx. 8.5 g/l transforming more than 85% of the total nitrogen into ammonia-nitrogen.

Based on simple dissociation-constants unrealistic high values of free ammonia are calculated. Taking ion-activities and measured concentrations of volatile fatty acids in the range of 5 to 25 g/l into account, the real niveau of free ammonia is in the range of 100-300 mg/l which corresponds perfect to the first values published in the context of ammonia-toxicity. In this contribution it is shown that high concentrations of ammonia are causing high concentrations of volatile fatty acids which act as acid counter-ion to the ammonium-ion and therefore are necessary to keep values of free ammonia (NH_3) low. Thus high concentrations of VFA are not an indicator for severe process instabilities in the context with high ammonia-concentrations. The process of anaerobic digestion of high nitrogen containing substrates may be described as a self-establishing equilibrium between the alkaline ammonia-ions (NH_4^+) and the acid group of volatile fatty acids. Both values have to be taken into consideration for a proper bench-mark of these fermentation parameters.

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8. Influence of operational conditions on the performance of a mesh filter activated sludge process

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Influence of operational conditions on the performance of a mesh filter activated sludge process

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Abstract

Recently, a new type of wastewater treatment system became the focus of scientific research, the mesh filter activated sludge system. It is a modification of the membrane bioreactor where a membrane filtration process serves to separate the sludge from the purified effluent. The difference is that a mesh filter is used instead of the membrane. Due to the much larger pore size of the mesh, the effluent is not of the same excellent quality as with membrane bioreactors. Nevertheless, it still resembles the quality of the now most widely used standard treatment system, where settling tanks are used to retain the activated sludge. At the same time, the new system features all the other advantages of membrane bioreactors including elevated sludge concentrations resulting in decreased volumina of basins and complete substitution of the settling tank. Therefore, this process presents a potential future alternative where a small footprint of the plant is required. However, so far only a few preliminary studies on this innovative process type have been done. In this paper, the effects of suspended solids concentration, flux rate as well as aeration rate on the effluent quality are discussed. Furthermore, the characteristic of the sludge floc was identified as a factor of vital importance. Therefore, another influencing parameter, the food to microorganism (F/M) ratio, which is known to have a significant effect on floc characteristics, was studied. The main result demonstrated that the process was very effective under most of the operation conditions. The suspended solids concentration in the effluent was below $12\,mg\,l^{-1}$, the average COD in the effluent was between 24 and $45\,mg\,l^{-1}$ and the BOD_5 was lower than $5\,mg\,l^{-1}$. High flux rates of up to $150\,l\,m^{-2}\,h^{-1}$ were also achieved.

Keywords: Activated sludge; Mesh filter; Wastewater treatment; Sludge floc; F/M ratio

1. Introduction

Membrane bioreactors are activated sludge processes where the secondary settling tank is substituted by a membrane filtration unit. The main advantages are the high sludge concentration in combination with an excellent effluent quality. In recent years, interest in this technology has considerably increased and several full-scale plants have been installed (Stephenson et al., 2000; Cicek, 2003; Frost&Sullivan, 2003). The driving force for the application of membrane bioreactors is, on the one hand, the small area requirement and, on the other hand, the disinfection of the effluent, which makes the membrane bioreactor a highly attractive system in cases where stringent standards have to be met, e.g. for discharge in bathing water or for water reuse (Guender, 2001; MUNLV NR, 2002).

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However, membrane bioreactors still need further improvement for a more widespread application. The main hindrances are the high costs of investment for the membrane modules and the elevated energy consumption (Guender, 2000, Gander et al., 2000). Especially for the extension of the capacity of existing wastewater treatment plants, it is often found that the advantage of the compactness of the process is of high interest whereas an improved effluent quality is not required and only standard criteria are stipulated. In this context, it should be suitable to substitute the membranes by a cheaper filtration technique which offers higher flux rates at lower transmembrane pressures. Keeping in mind that the activated sludge appears as flocs, a coarser filter material can be used to separate the activated sludge from the treated wastewater. So far, mesh filtration systems have been mainly used for the polishing of secondary wastewater effluents (Grabbe et al., 1998). Only within the last few years has the idea of direct sludge separation been studied (Kiso et al., 2000, Alavi Moghaddam et al., 2002a, b, Seo et al., 2002).

Preliminary investigations in our laboratory showed that the quality of the effluent varies under different operation conditions. The critical issue is the formation of a sludge layer on the mesh surface which serves as the actual filter membrane. Important parameters of influence are shear stress at the filter surface, suction rate and suspended solids (SS) concentration. Also the nature of the activated sludge and its floc structure affect the proper establishment of a secondary filter and hence the quality of the effluent. It is known that the substrate availability, i.e. the food to microorganism (F/M) ratio, has a specific influence on sludge characteristics and floc size distribution (Barbusiński and Kościelniak, 1995). Therefore, in this study, the effect of the above-mentioned parameters including various F/M ratios on the process performance was investigated.

2. Experimental

2.1. Mesh filter bioreactor

A schematic layout of the bioreactor with its submerged mesh filter is presented in Fig. 1.

The working volume of the reactor was 30 l. It contained a flat sheet type filter module made from a woven nylon fabric of 30 μm mesh width which was stretched over a rectangular frame. The active filter area had a dimension of two times 220 × 220 mm at a distance of 15 mm. The resulting effective filter area was 968 cm^2 or roughly 0.1 m^2. An aeration unit was placed below the filter module serving both for aeration of the activated sludge and generation of turbulence to minimise the filter layer on the mesh filter. The aeration unit was a standard membrane diffuser of 175 mm diameter (Sanitaire, membrane disc diffuser). The flow rate near the mesh filter was measured by a propeller flow meter (Schildknecht, Miniwater 2 Micro) which was inserted at several points in the reactor close to the mesh surface.

A temperature control system was not installed and temperature fluctuated between 14 and 17 °C according to ambient conditions.

A peristaltic pump (Watson Marlow, 505 U) was used to generate the necessary trans-filter pressure by suction of the filtrate. The trans-filter pressure was continuously monitored by a pressure inducer (Wika, −1 to 0 bar) in the suction line. The gained values were automatically

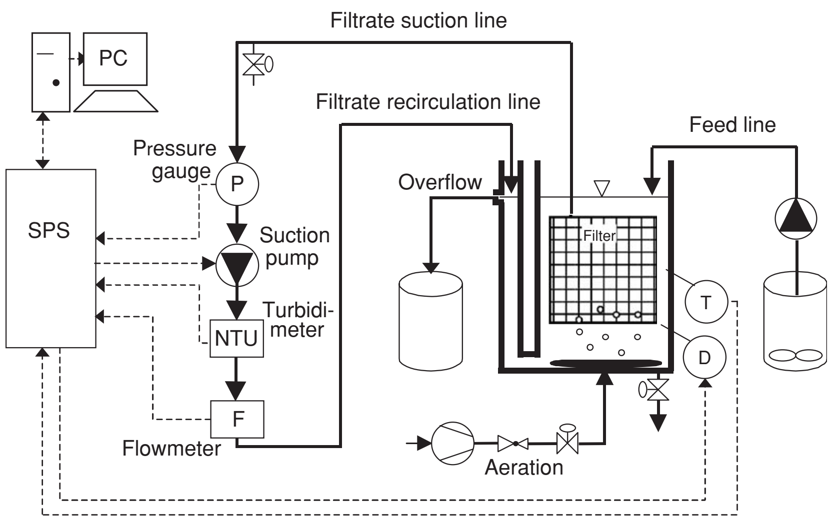

Fig. 1. Schematic layout of the reactor set-up.

corrected for the hydrostatic pressure difference between the water level in the reactor and the measuring point. The filter was operated at a preset flow rate. It was controlled by a flow meter (McMillan, 100-6 flow sensor) adjusting the rotation speed of the pump.

To maintain a constant filter flow independently of the wastewater feeding rate, part of the filtrate was recycled into the reactor. For this purpose, the filtrate was fed into a small tubular vessel connected to the side of the bioreactor. In the upper part of the tube, a small outflow was located which determined the surface level within the reactor. Using this set-up ensured that the volume within the bioreactor was always kept at the same level.

The suction line was equipped with an in-line turbidimeter (WTW, Turb 555) for the monitoring of the turbidity of the filtrate.

All data were fed into an automatic controller (Mitsubishi, FX2N) which was connected to a PC for parameter setting and data storage.

2.1.1. Analytics

2.1.1.1. Influent and effluent composition. Beside the turbidity the typical wastewater parameters, COD, BOD_5, NH_4-N, NO_3-N and SS were measured in the influent and effluent. All analyses were done using standard analytical procedures or commercially available kuvette test kits (Dr. Lange). Turbidity and SS concentration in the effluent showed a very high linear correlation, with 1 turbidity unit corresponding to 1.46 mg SS l^{-1} (Fig. 2). This calculation was used to convert on line turbidity data into SS concentration.

2.1.1.2. Sludge characteristics. Mixed liquor suspended solids (MLSS) were regularly monitored to adjust the sludge concentration to a preset level. To describe the morphology of the activated sludge characteristics, microscopic investigation was used. The floc size distribution was documented according to the widely used scheme developed by Eikelboom and van Buijsen (1983). Three different groups were distinguished: big flocs with a diameter >500 μm, flocs of intermediate size, 150–500 μm, and small sludge flocs <150 μm.

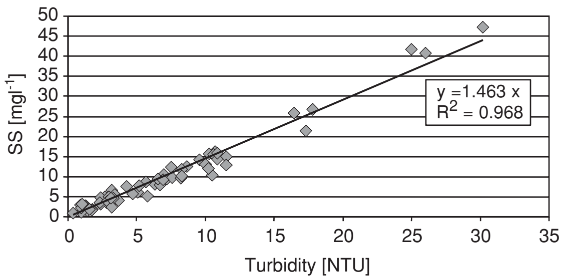

Fig. 2. Correlation between turbidity and SS in the effluent.

2.1.1.3. Wastewater. Raw wastewater was derived from a local municipal sewage treatment plant. It was sampled once or two times per week after screening and grid separation. For each run, the feeding rate was adjusted according to the actual BOD concentration determined. The typical composition of the wastewater is presented in Table 1.

3. Results and discussion

3.1. General performance of the mesh filter

3.1.1. Start-up phase

In all experiments, the filter exhibited typical behaviour in the start-up phase. A representative graph is shown in Fig. 3. Initially, the SS concentration was relatively high. With time, it dropped gradually presumably due to the build-up of the secondary filter layer. Within 12–24 h, the SS concentration stabilised at a low level. The trans-filter pressure did not show a change in that period.

As proven by other experiments, the slightly higher SS concentration in the start-up phase can be even further reduced if the selected flow rate is not fully applied from the beginning, but gradually increased.

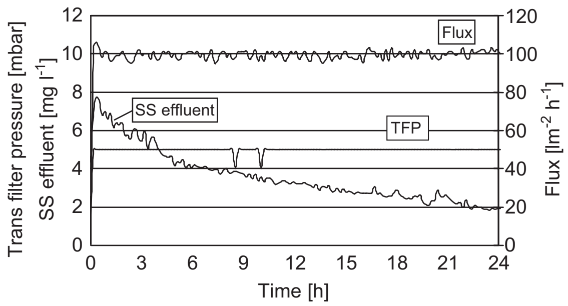

Fig. 3. Example of the general behaviour of the mesh filter in the start-up phase. Data from run no. 17 (Table 3): flux 100 l m^{-2} h^{-1}, MLSS 4.0 g l^{-1}, F/M 0.3 g BOD g^{-1} MLSS d^{-1}.

Table 1
Composition of the wastewater

Parameter	COD (mg l^{-1})	BOD (mg l^{-1})	SS (mg l^{-1})	N-org (mg l^{-1})	NH_4-N (mg l^{-1})	Total-P (mg l^{-1})	PO_4-P ortho (mg l^{-1})
	270–570	130–310	112–234	30–76	17–49	4.9–12.4	1.9–5.5

3.1.2. Filter blocking

The trans-filter pressure was only about 3–10 mbar and was raised very slowly. Depending on the operation conditions after a filtration period between a few days and sometimes up to 2–3 weeks, trans-filter pressure increased suddenly, often within minutes, to values as high as 100 mbar and almost complete blocking of the filter occurred. At this point, the filtration was automatically stopped. In general, there was no obvious indication of impending filter blocking. This general pattern was observed in almost all runs independently of the chosen filtration rate, the sludge concentration and the aeration rate. Typical examples are presented in Fig. 4. A different behaviour was only observed in the two runs where a higher F/M ratio of 0.18 and 0.3, respectively, was applied. In these cases, a gradual increase of trans-filter pressure was observed (Fig. 5). Intensive flushing on air of the blocked filter was sufficient to fully re-establish filter performance.

In similar studies on mesh filter systems, both patterns of filter behaviour were observed. Some investigators report gradual increase of trans-filter pressure (Kiso et al., 2000, Wu et al., 2003), whereas Fan and Huang (2002) point out stable filtration periods and a steep increase of trans-filter pressure at the end.

In some of the experiments described here, periodical inspection of the mesh filter by light-optical and to a smaller extent by scanning electronic microscopy was done. However, these investigations did not provide a consistent picture of the nature of the blocking mechanism and the issue remains subject to future investigations.

3.2. Effect of flux, aeration rate and suspended solids

As a first setting, two different suction rates corresponding to a constant flux of 50 and 150 $l\,m^{-2}\,h^{-1}$ were investigated. However, in wastewater treatment plants, the flow rate shows a typical pattern of daily fluctuation. Therefore, in an additional experiment, a daily flow variation was simulated by gradually increasing the flux from 50 to 150 and back to 50 $l\,m^{-2}\,h^{-1}$ in a 24 h cycle.

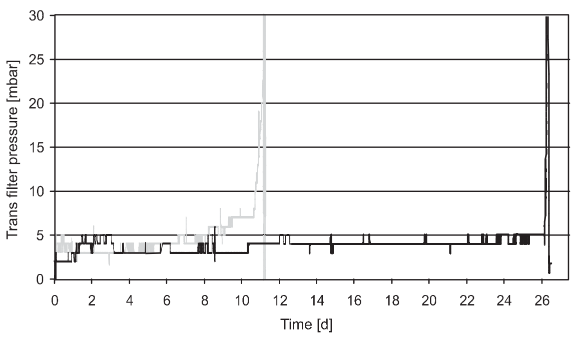

Fig. 4. Typical examples for the general behaviour of trans-filter pressure. Data from run no. 15 (grey line; Table 3): flux 100 $l\,m^{-2}\,h^{-1}$, MLSS 4.0 $g\,l^{-1}$, F/M 0.06 g BOD g^{-1} MLSS d^{-1}; data from run no. 19 (black line; Table 3): flux 100 $l\,m^{-2}\,h^{-1}$, MLSS 4.0 $g\,l^{-1}$, F/M 0.06 g BOD g^{-1} MLSS d^{-1}.

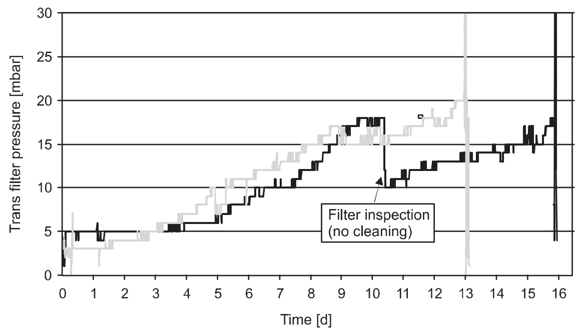

Fig. 5. Raise of trans-filter pressure at higher F/M ratios. Data from run no. 16 (grey line; Table 3): flux 100 $l\,m^{-2}\,h^{-1}$, MLSS 4.0 $g\,l^{-1}$, F/M 0.18 g BOD g^{-1} MLSS d^{-1}; data from run no. 17 (black line; Table 3): flux 100 $l\,m^{-2}\,h^{-1}$, MLSS 4.0 $g\,l^{-1}$, F/M 0.30 g BOD g^{-1} MLSS d^{-1}.

The second investigated parameter was the aeration rate. As described earlier, the aeration fulfils two tasks: on the one hand, the supply of oxygen to the activated sludge and, on the other hand, the generation of turbulence to keep the secondary layer on the mesh filter at a constant thickness. To see the effect, the tests were run at two different aeration rates, 2 and 8 $l_n\,min^{-1}$. In both cases, the air supply was sufficient to maintain an oxygen concentration well above 2 $mg\,l^{-1}$. The flow velocity near the mesh surface was approximately 0.25–0.3 $m\,s^{-1}$ for 8 $l_n\,min^{-1}$ and only 0.06–0.08 for an aeration rate of 2 $l_n\,min^{-1}$.

Furthermore, all experiments were repeated at two levels of SS concentration, 4.0 ($\pm$0.7) and 7 ($\pm$1) $mg\,l^{-1}$. In the last run of this first series, an experiment with a constant flow rate of 200 $l\,m^{-2}\,h^{-1}$ was performed.

The complete list of settings with the corresponding parameters is given in Table 2. Each run was performed for a duration of 1 week or, in certain cases, until blocking of the filter occurred.

Fig. 6 shows the acquired results in terms of SS in the effluent. At the low sludge concentration, an increase of the flux rate had a small effect on the SS concentration in the effluent. The highest concentration of SS was found in the experiments with varying flux rates. Compared to runs at the sludge concentration of 4.0 $g\,l^{-1}$ MLSS, almost all corresponding experiments at a sludge concentration of 7 $g\,l^{-1}$ showed higher SS concentrations in the effluent. Interestingly, in these experiments, the influence of flux rate on the SS concentration did not follow any evident pattern.

It was also generally observed that the higher aeration rate resulted in an elevated SS concentration.

Table 2
Operational parameters of the mesh bioreactor for the experiments with different sludge loading rates

Run no.	Flux rate of filtrate ($l\,m^{-2}\,h^{-1}$)	Aeration rate ($l_n\,min^{-1}$)	Sludge concentration (MLSS) ($g\,l^{-1}$)	F/M ratio (g $BOD\,g^{-1}$ $MLSS\,d^{-1}$)	Volumetric loading rate B_R (g $BOD\,l^{-1}\,d^{-1}$)	Average hydraulic retention time (d)	Average COD in the effluent	Days of operation until filter blocking (d)
1	50	2	4.0	0.05	0.200	1.36	36.4	—[a]
2	50	8	4.0	0.05	0.200	0.88	25.6	—[a]
3	150	2	4.0	0.05	0.200	0.81	36.4	—[a]
4	150	8	4.0	0.05	0.200	0.87	32.5	—[a]
5	50–150	2	4.0	0.05	0.200	0.89	47.0	—[a]
6	50–150	8	4.0	0.05	0.200	1.00	31.9	—[a]
7	50	2	7.0	0.05	0.350	0.61	24.6	3.5
8	50	8	7.0	0.05	0.350	0.67	45.1	3.5
9	150	2	7.0	0.05	0.350	0.51	24.6	4.5
10	150	8	7.0	0.05	0.350	0.63	23.8	3.5
11	50–150	2	7.0	0.05	0.350	0.52	23.8	4.5
12	50–150	8	7.0	0.05	0.350	0.55	35.7	6.0
13	200	8	4.0	0.05	0.200	1.2	63.5	3.0

[a]No filter blocking within the duration of the run of 1 week.

The findings are consistent with the results reported by Ozaki and Yamamoto (2001). They investigated sludge layer formation on a flat sheet membrane in an MBR under different hydraulic conditions and at three different MLSS concentrations in the range of 0.8–6.0 $g\,l^{-1}$. According to their findings, the sludge accumulation on the membrane surface was only weakly dependent on MLSS concentration whereas aeration intensity played a significant role. For the results presented here, it can be deduced that higher MLSS rates did not significantly increase cake layer thickness. Because of the generally increased number of flocs at higher MLSS concentrations, a bigger amount of the small flocs passes through the cake layer resulting in an elevated SS concentration in the effluent. On the other hand, the higher shear stress at increased aeration reduced the thickness of the secondary filter layer, which made the retention of SS less effective.

An exception was experiment no. 5 which showed an atypically high SS concentration. The explanation for the higher SS concentration in this run is that the reactor temperature dropped to a value as low as 8 °C due to a failure of the laboratory heating systems. This is also the reason for the slightly elevated COD concentration in the effluent.

The most obvious effect was that the elevated sludge concentration had a significant impact on the blocking behaviour of the mesh filter. In this set of experiments, all runs were stopped after a maximum duration of 1 week, a time where at the low sludge levels no indication of filter blocking was detectable. From previous experiments, it was known that the mean operation time for low sludge concentrations was 2–3 weeks before filter blocking occurs. In contrast, in all the runs at the higher sludge level after a typical operation period of 3.5–4.5 d, filter blocking was observed. It was expected that the higher aeration rate has a beneficiary effect on the filtration period until blocking, but the filtration time was independent of the aeration level.

As previously explained, the influence of the tested flow rates was generally small. Therefore, the last test run with a flux of 200 $l\,m^{-2}\,h^{-1}$ was conducted to see where the limit lies. This final run was clearly beyond the capacity of the mesh filter. It was not possible to achieve a constant filtrate quality. An average SS concentration of 39 $mg\,l^{-1}$ with maximum values over 100 $mg\,l^{-1}$ was measured. To exclude the possibility of undetected filter leakage, a repetition of the run was conducted and similar results were acquired.

4. Effect of F/M ratio on effluent quality

These experiments were carried out at a constant sludge concentration in the range of 3.5–4.5 $g\,l^{-1}$ and an

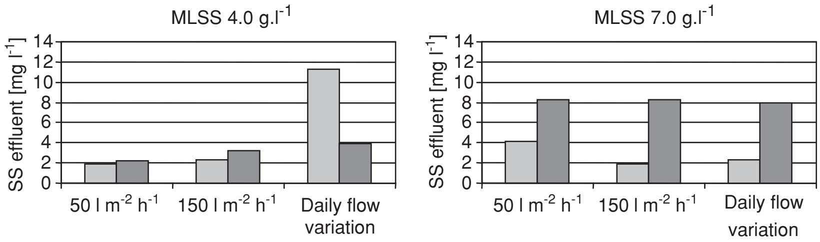

Fig. 6. Comparison of SS concentration in the effluent at different operation conditions as listed in Table 2. (a) (left) Results at an activated sludge concentration of $4.0\,g\,l^{-1}$ MLSS at different flux rates and aeration rates of $2\,l_n\,min^{-1}$ (light bars) and $8\,l_n\,min^{-1}$ (dark bars). (b) (right) Corresponding results at a sludge concentration of $7\,g\,l^{-1}$.

Table 3
Operational parameters of the system for the experiments with different sludge loading rates

Run no.	Flow rate of filtrate ($l\,m^{-2}\,h^{-1}$)	F/M ratio (g BOD g^{-1} MLSS d^{-1})	Volumetric loading rate B_R (g BOD $l^{-1}\,d^{-1}$)	Average hydraulic retention time (d)	Average COD in the effluent ($mg\,l^{-1}$)	Days of operation until filter blocking (d)
14	100	0.03	0.120	1.42	39.3	—[a]
15	100	0.06	0.240	0.56	26.0	11.5
16	100	0.18	0.720	0.21	39.4	13
17	100	0.30	1.200	0.15	37.0	16
18	100	0.0	0.0	—	n.d.	—[a]
19	100	0.06	0.240	0.76	25.5	26

[a]No blocking of the filter within the operation period of 2 weeks.

aeration rate of $8\,l_n\,min^{-1}$. The F/M ratio was varied between 0.0 and 0.3 g $BOD_5\,g^{-1}$ MLSS d^{-1}. A constant mesh filter flow rate of $100\,l\,m^{-2}\,h^{-1}$ was applied by partial recycling of the filtrate as described earlier. An overview of operational parameters is given in Table 3.

In each experiment, an approximately three-fold volume change was considered as adaptation period to the modified F/M ratio. The corresponding data were not taken into account. Each experiment was operated for about 2 weeks or until filter blocking occurred.

At the end, the setting with an F/M ratio of 0.06 was repeated to see whether results are reproducible. In this case, the run was not stopped after 2 weeks but continued until day 26 when filter blocking was observed.

As expected, the F/M ratio had a critical influence on the floc characteristics. In accordance with observations in a membrane bioreactor (Huang et al., 2001), the higher the F/M ratio, the higher the number of intermediate and large size flocs and conversely the fewer small flocs observed. The consequences are best illustrated in Fig. 7. It is obvious that bigger flocs are associated with a better effluent quality in terms of SS content. Obviously, they bridge the gaps between the mesh grid, thereby contributing to the formation of a secondary layer on the mesh which serves as the actual filter. In contrast, when only smaller flocs are present,

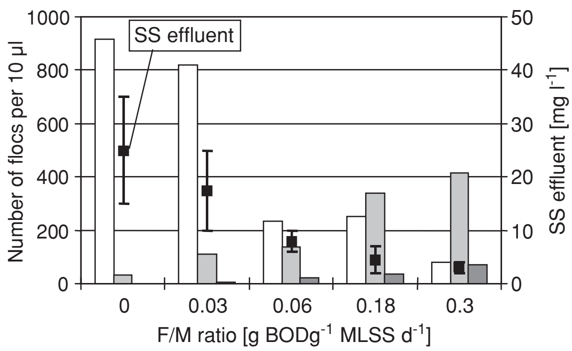

Fig. 7. Correlation of sludge characteristics in terms of floc size distribution and SS concentration in the effluent. Black dots with error bars: SS effluent; bars: light, flocs $<150\,\mu m$; grey, flocs 150–500 µm; black, flocs $>500\,\mu m$.

they are only partly held back and pass through the grid to some extent. The best results were achieved at the highest F/M ratio of 0.18 and 0.3 g $BOD_5\,g^{-1}$ MLSS d^{-1} with high-quality effluent <7 mg SS l^{-1}. A significant drop in quality was observed only at very low F/M ratios. Somehow comparable results were obtained in experiments by Alavi Moghaddam et al. (2002a, b) with a coarse pore filtration activated sludge process. At F/M ratios of 0.18 and 0.1 g TOC g^{-1} MLSS d^{-1}, they observed an average SS concentration of 1.7 and 2.9 mg l^{-1} in the effluent. In contrast, the SS concentration at an F/M ratio of only 0.05 was highly fluctuating with a mean value of 23.5 mg l^{-1}. However, their results were influenced by the fact that lower F/M ratios were achieved by an increase in MLSS concentration in the bioreactor while the substrate feeding rate was kept constant. Therefore, worsening of the effluent quality might also partly be attributed to higher MLSS concentrations, which were 1.5, 2.6 and finally 5.2 g l^{-1} towards lower F/M ratios.

5. Removal capacity and overall process performance

The EC directive concerning urban wastewater treatment (Directive 91/271/EC) establishes a maximum value of 35 mg l^{-1} total SS with a minimum percentage of reduction of 90%, as well as a maximum COD concentration of 125 mg l^{-1} with a minimum percentage of reduction of 75% (for treatment plants $>$10 000 PE). For well-functioning standard activated sludge processes, effluent concentrations of 10–15 mg l^{-1} SS and 40–50 COD are given (ATV-DVWK, 2000). In this respect, the achieved COD, BOD_5 and SS removal was more than satisfactory. At appropriate operation conditions, the SS concentration in the effluent was below 12 mg l^{-1}. The average COD in the effluent was between 24 and 45 mg l^{-1} and the BOD_5 was always lower than 5 mg l^{-1}.

It was also demonstrated that the effluent quality is highly satisfactory in a wide range of F/M ratios from 0.06 to 0.3 g BOD g^{-1} MLSS d^{-1}: typical F/M ratios for activated sludge processes are 0.3 for extensive carbon removal, 0.15 to achieve nitrification/denitrification and 0.05 for aerobic sludge stabilisation.

High flux rates ranging from 50 to 150 l m^{-2} h^{-1} were successfully applied. For a similar type of reactor using a 100 μm nylon mesh, Kiso et al. (2000) achieved flux rates of 21–32 l m^{-2} h^{-1}. Fan and Huang (2002) applied 15–33 l m^{-2} h^{-1} to a polyester mesh of the same pore size. Nonwoven fabrics with an irregular pore structure were also investigated for direct sludge separation. Alavi Moghaddam et al. (2002a, b) reported stable flux rates of 42 l m^{-2} h^{-1} (nonwoven fabric made from polyester) and Seo et al. (2002) reported 17 l m^{-2} h^{-1} (nonwoven fabric, polypropylene). On the other hand, the standard design flux rate of submerged membranes is 25–30 l m^{-2} h^{-1} (ATV-DVWK, 2000).

However, it was also observed that filter blocking occurs, with operation periods of only half a week at elevated activated sludge concentrations. Kiso et al. (2000) and Alavi Moghaddam et al. (2002a) report permanent filtration without filter blocking for several months at the above-cited flux rates in the range of 20–40 l m^{-2} h^{-1}. Nevertheless, they stress filter blocking as a critical issue. Fan and Huang (2002) found a strong dependency on flux rate and in their experiments filtration periods before blocking decreased from 7d at a flux of 15 l m^{-2} h^{-1} to less than 0.5 d at a flux of 33 l m^{-2} h^{-1}.

Therefore, a problem still to resolve is the periodical cleaning of the mesh filter. In membrane bioreactors, the submerged membranes are often operated in a discontinuous way. Following a particular operation period, backwashing of the membrane or relaxation, that is, a stop of permeate production to allow removal of the filter cake by the cross-flow, is performed. This process is cyclically repeated typically at intervals of about 10 min (Albasi et al., 2002). In these experiments, no such procedure was applied. Ongoing investigations are focusing on the development of appropriate cleaning techniques. A potential solution is intermittent air sparging which was successfully applied by Kiso et al. (2000) to continuously operate a mesh filter bioreactor at MLSS concentrations of 4.3–7.0 g l^{-1} over a period of several months. As well Fan and Huang (2002) claim that air sparging was sufficient to clean and restore the clogged mesh filters.

6. Conclusions

The suitability of a mesh filter as a sludge separation device in municipal wastewater treatment was investigated. Using a filter of 30 μm mesh size, it was demonstrated that the appropriate development of a secondary filter layer is of critical importance to effluent quality. In general, it was possible to fulfil all standards in terms of COD, BOD and SS removal. High flux rates from 50 to 150 l m^{-2} h^{-1} were achieved. At the same time, the pressure loss was very low.

In conclusion, the mesh filter bioreactors revealed considerable potential as a wastewater treatment process where a small area requirement is a high priority. Similar advantages to membrane bioreactors with regard to elevated sludge concentration and volumetric loading rates are addressed. At the same time, an effluent quality equal to well-operated standard activated sludge systems can be achieved.

Nevertheless, technical challenges for the practical realisation remain, in particular with regard to periodical filter cleaning. It is our hope that the acquired findings stimulate future research and development of this new and attractive wastewater treatment process.

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Further reading

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