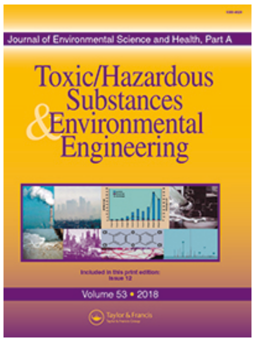

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Environmental impact of biogas: A short review of current knowledge

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ABSTRACT
The social acceptance of biogas is often hampered by environmental and health concerns. In this study, the current knowledge about the impact of biogas technology is presented and discussed. The survey reports the emission rate estimates of the main greenhouse gases (GHG), namely CO_2, CH_4 and N_2O, according to several case studies conducted over the world. Direct emissions of gaseous pollutants are then discussed, with a focus on nitrogen oxides (NO_x); evidences of the importance of suitable biomass and digestate storages are also reported. The current knowledge on the environmental impact induced by final use of digestate is critically discussed, considering both soil fertility and nitrogen release into atmosphere and groundwater; several case studies are reported, showing the importance of NH_3 emissions with regards to secondary aerosol formation. The biogas upgrading to biomethane is also included in the study: with this regard, the methane slip in the off-gas can significantly reduce the environmental benefits.

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Air quality; anaerobic digestion; biogas; digestate; renewable energy; secondary aerosol; waste management

Introduction

The environmental benefits of biogas technology are often highlighted, as a valid and sustainable alternative to fossil fuels.[1] Together with the reduction of greenhouse gas (GHG) emissions, biogas can enhance energy security, thanks to its high energetic potential.[2–4] As a renewable energy source, it allows exploiting agricultural and zootechnical byproducts and municipal wastes, with a lower impact on air quality when compared to combustion-based strategies for these biomasses.[5–7] Furthermore, while ashes from combustion find scarce agronomic applications,[8,9] the by-product of anaerobic digestion, i.e. digestate, looks as a reliable material for agricultural uses.[10] Another important advantage of biogas technology is its easy scalability, allowing exploiting the energetic potential of decentralized biomass sources.[11,12] Finally, biogas can be upgraded to biomethane, suitably used as a vehicle fuel, or injected into national natural gas grids.[13,14]

The energy potential of biogas is reported in Figure 1, based on data from the World Bioenergy Association.[15] For Europe, China and USA, data are detailed in terms of the following sources: manure, agriculture residues, energy crops, organic fraction of municipal solid waste (MSW), agro-industry waste and sewage sludge. For the total world biogas potential, data are only divided into waste (i.e. organic fraction of MSW, agro-industry waste and sewage sludge) and agricultural byproducts (i.e. manure, agriculture residues and energy crops).

In spite of the above cited advantages, social opposition is often observed towards biogas plants, generally based on concerns about environmental and health issues.[16] The frequency on which these opposition phenomena are observed depends on different factors, including the inclusion strategies and the considered country.[17,18] In order to overcome social and cultural barriers hampering a wider diffusion of biogas, the accurate and complete evaluation of the environmental impact of these processes remains an issue of high scientific and technical relevance. The aim of this work is to report an updated state of the art of current knowledge about the environmental impact of biogas and biomethane.

Greenhouse gas emissions

A main objective of biogas industry is the reduction of fossil fuel consumption, with the final goal of mitigating global warming. However, anaerobic digestion is associated to the production of several greenhouse gases, namely carbon dioxide, methane and nitrous oxide. As a consequence, dedicated measures should be taken in order to reduce these emissions. According to Hijazi,[19] the main measures to improve the global warming reduction potential of biogas plants are: to use a flare avoiding methane discharge, to cover tanks, to enhance the efficiency of combined heat and power (CHP) units, to improve the electric power utilisation strategy, to exploit as much thermal energy as possible, to avoid leakages. Similar conclusions were obtained by Buratti and co-workers[20] for the specific case study of cereal crops in Umbria, Italy. Biomethane chain exceeds the minimum value of GHG saving (35%) mainly due to the open storage of digestate; usual practices to improve GHG reduction (up to 68.9%) include using heat and electricity produced by the biogas CHP plant, and covering digestate storage tanks.

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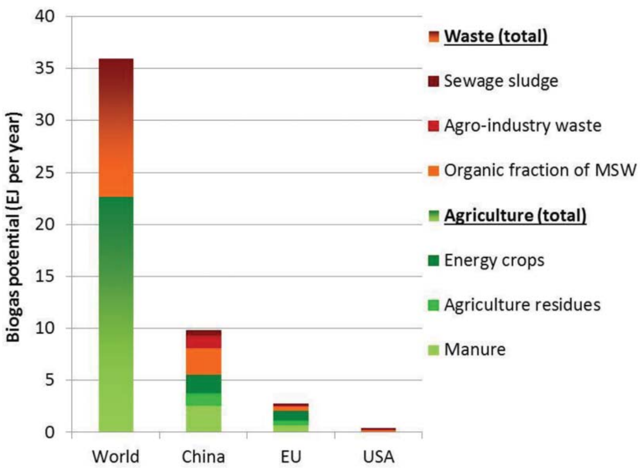

Figure 1. Energy potential of biogas.

The impact induced by biogas plants on global warming needs to be studied case by case. Bachmaier and co-workers[21] calculated the GHG impact of ten agricultural biogas plants. GHG emissions coming from electricity production in the investigated biogas plants ranged from −85 to 251 g CO_2-eq/kWh_{el}, and the GHG saving was 2.31 – 3.16 kWh_{fossil}/kWh_{el}. The results obtained also highlighted that reliable estimates of GHG emissions in the case of electricity production from biogas can be only made on the basis of individual monitoring data, for instance: reduction of direct methane emission and leakage, exploiting of heat obtained from cogeneration, amount and nature of input material, nitrous oxide emission (e.g. from energy crop cultivation) and digestate management. Battini and co-workers,[22] in a case study of an intensive dairy farm situated in the Po valley (Italy), calculated a GHG emission reduction due to anaerobic digestion ranging between −23.7% and −36.5%, depending on digestate management. In a Finnish case study,[23] the GHG release reduction was estimated equal to 177.0, 87.7 and 125.6 Mg of CO_2 eq. yr^{-1} for dairy cow, sow and pig farms, respectively. Optimizing all process parameters looks important with regard to final environmental impact: for instance, a specific case study on wastewater treatment showed that the process optimization could result into the emission abatement equal to 1,103 kg CO_2 eq/d for N_2O, 256 kg eq/d for CO_2 and 87 kg CO_2 eq/d for CH_4.[24]

Carbon dioxide emissions

Harmful compounds and air contaminants are introduced into the environment during biogas production and use through both combustion processes and diffusive emissions. Considering carbon dioxide, combustion of biogas leads to efficient methane oxidation and conversion to CO_2, with a rate of 83.6 kg per GJ (based on a biogas with 65% CH_4 and 35% CO_2[25]). Other releases of this contaminant are related to transport and storage of biomass, as well as digestate use. In the case of both biogas combustion and biomass/digestate emission, CO_2 is considered as biogenic and calculated neutral with regards to the impact on climate. Taking into account the reduction of fossil fuel, it can be demonstrated that biogas production leads globally to mitigation of anthropogenic greenhouse impact of the environment. Poeschl and co-workers[26] have investigated the CO_2 emissions associated to biogas production from several feedstocks, and the relative contribution of feedstock supply, biogas plant operation and infrastructure, biogas utilization and digestate management. According to this study, biogas use gives rise to a negative CO_2 balance because CO_2 caption results every time higher, in absolute values, than positive emissions from feedstock supply and biogas plant operation. As expected, biogas production from byproducts (e.g. from food residues, pomace, slaughter waste, cattle manure, etc.) is a more sustainable approach than energy crops utilization such as whole-wheat plant silage. Besides, digestate management provides significant contributions to total emission reduction in the case of specific feedstock such as municipal solid waste. A dedicated section of this study will below discuss the impact of digestate in full details, in paragraph 5.

Methane emissions

Methane released by biogas processes is not considered relevant for health issues: though exposure to hydrocarbon mixtures can have some adverse effects on humans,[27] no evidence exists of relevant interactions between methane and biologic systems.[28] However, methane is a greenhouse gas whose global warming power is estimated to be 28–36 times higher than CO_2 over 100 years: as such, it is the second major component among anthropogenic greenhouse chemicals.[29] Hence, in evaluating the impact of biogas industry on climate change, methane emissions are a point of primary importance. Methane can be released during biogas incomplete combustion; however a strong contribution to this contaminant comes out from diffusive emission related to biomass storage and digestate management. On the other hand, other biomass management strategies must be taken into account to abate emissions related to biogenic methane. In the above mentioned study of Poeschl and co-workers,[26] methane emissions were also discussed; in all investigated cases, the emission rates were below 5 g kg^{-1}. Considering cattle manure, important reductions in methane emission are related to digestate processing and handling, since this kind of biomass is characterized by high methane emission rate when spread in the field without any pre-treatment.

Nitrous oxide

Besides CO_2 and CH_4, nitrous oxide (N_2O) is another important GHG: Due to its high greenhouse effect potential, N_2O emissions from biogas production processes can result into a significant contribution to global warming budget.[30,31] The relative impact of nitrous oxide mostly depends on the chosen climate metrics: indeed, N_2O impact can even exceed those of CO_2 and CH_4, when the considered metric is Global Temperature change Potential with a time horizon of 100 years (namely GTP-100).[32]

Total GHG emission for energy production from biogas are generally calculated in a range between 0.10 and 0.40 kg CO2-eq/kWh_{el}, which is for instance 22–75% less than GHG emissions caused by the present energy mix in Germany.[33] The wide uncertainty about the estimates of global warming mitigation potential depends on N_2O emission rate assessment as well as on storage and use as a fertilizer of digestate, as discussed in paragraphs below.

Gaseous pollutants from biogas combustion

Along GHG reduction benefits, it must be considered that biogas combustion is associated to release of pollutants in the atmosphere; therefore, the correct assessment of these emissions is a key point in social acceptance of this technology. A summary of emission factors for the main gaseous pollutants are reported in Table 1.

Carbon monoxide (CO) is produced in all oxidation processes of carbon containing materials, and is an important by-product of incomplete combustion of biogas. Methane emission rates are 0.74 and 8.46 and g CO per Nm^{-3} CH_4 for flaring and CHP, respectively.[34] CO emissions related to energy production are estimated in a range between 80 and 265 mg CO MJ^{-1}, depending on the plant efficiency.[35]

Sulphur dioxide (SO_2) emissions from biogas plants manly depend on the desulphurization degree of the introduced biogas. The SO_2 emission rate of a CHP biogas plant is estimated to lie in the range 19.2–25 mg MJ^{-1}.[25] The UK National Society for Clean Air (NSCA) estimates an emission factor of 80 and 100 $g_{SO2}/tonn_{waste}$ for flaring and CHP, respectively.[36] The relatively high SO_2 concentrations in the proximity of biogas plants can depend on different reasons, e.g.: direct emission from biogas combustion, H_2S oxidation from diffusive emissions, and diesel truck exhausts.[37]

Emissions of NO_x are one of the most critical point with regard to environmental impact of biogas plants.[38] According to Kristensen and co-workers,[35] the NO_x emission level of biogas is, in general, higher than for natural gas engines: the averaged aggregated emission factor is 540 g NO_x GJ^{-1}, which is more than three times the rate from natural gas engines. When emission factor is reported to methane consumption, an emission factor of 0.63 and 11.6 $g_{NOx}/Nm^3{}_{CH4}$ can be assumed for flaring and CHP, respectively.[34] The importance of controlling this pollutant is demonstrated by several case studies. For instance, Battini and co-workers[22] in the above mentioned case study of an intensive dairy farm situated in the Po valley (Italy) reported a low enhancement in acidification (5.5–6.1%), particulate matter emissions (0.7–1.4%) and eutrophication (+0.8%), while on the other hand a significant enhancement in photochemical ozone formation potential (41.6–42.3%) was calculated. In another case study, Carreras-Sospedra and co-workers[39] estimated a potential enhancement of up to 10% of NO_x emission in 2020 in California (US); nevertheless, their study included both biogas and biomass burning. Indeed, the lower emissions of methane from storage and the credits from substituted electricity are not enough to compensate the increase in NO_x emissions from the biogas combustion.

Biogas is a gaseous fuel rich in volatile organic compounds (VOCs), compared to natural gas: indeed, VOCs concentration normally ranges between 5 and 500 mg/Nm^3, and in some cases up to 1700 mg/Nm^3 were observed.[40,41] Generally, only non-methane volatile organic compounds (NMVOC) are considered in these studies. If combustion is assumed to reduce VOCs concentration of 99%,[42] VOCs emission from biogas combustion are in general lower, compared to liquid and solid biofuels. However, a specific critical issue can be highlighted for formaldehyde. In a case study conducted on anaerobic waste treatment plants in Barcelona (Spain), VOC emission factors was in the range 0.9 ± 0.3 g s^{-1}, contributing for 0.3–0.9% of total VOCs in the area. On the other hand, formaldehyde emission factors from biogas engines were found between 0.2 and 3.0 mg s^{-1}, resulting in a ~2% contribution to the total.[43] It is important to remark that a similar emission pattern is observed for natural gas: indeed, formaldehyde is a by-product of methane oxidation. Compared to natural gas, emissions of VOCs are 40% lower in biogas engines, while formaldehyde emissions are slightly lower and higher aldehydes (present in natural gas due to the presence of higher hydrocarbons) are almost absent.[35]

Noticeably, fuel-cycle emissions can be strongly influenced by the raw materials. For instance, CO_2, CO, NO_x, hydrocarbons and particles may differ by a factor of 3–4 between ley crops, straw, sugar beet byproducts, liquid manure, food industry waste and municipal solid waste. On the other hand, differences by a factor of up to 11 can be observed in SO_2 emissions, due to the high variability of H_2S and organic sulphur compounds in the produced biogas.[44]

Impact of feedstock and digestate storage and treatment

In the biogas combustion management, feedstock and digestate storage and treatments can be the most important processes to achieve the global warming benefits of biogas production processes. Indeed, the impact of a biogas plant on GHG emission is heavily influenced by feedstock storage: most of N_2O can be abated when a closed storage is used for manure and co-digestion feeding.[45]

Emissions from uncovered biomass storage have also been identified as the main ammonia source along the whole biogas production chain,[46] and closed storage is strongly advised.

In a specific French case study of anaerobic digestion and composting plant for municipal solid waste, Beylot and co-workers[38] have identified four conditions for process operation, which highly influence the impact of the whole plant; they are: (i) the features of degradation of the fermentable fraction; (ii) the collection efficiency of gas streams released by biological operations; (iii) the abatement effectiveness of collected pollutants; and (iv) NO_x emission rate from

Table 1. Emission factors of biogas plants operating direct biogas combustion.

Pollutant	Emission factor (g GJ^{-1})	Source
Carbon monoxide (CO)	310	Nielsen et al.,[25]
	256	Kristensen et al.,[35]
Sulphur dioxide (SO_2)	25	Nielsen et al.,[25]
Nitrogen oxides (NO_x)	202	Nielsen et al.,[25]
	540	Kristensen et al.,[35]
Non-methane volatile organic compounds (NMVOC)	10	Nielsen et al.,[25]
	21.15	Kristensen et al.,[35]
Formaldehyde (CH_2O)	8.7	Nielsen et al.,[25]
	14	Kristensen et al.,[35]

biogas combustion. The importance of digestate storage step has been highlighted by Battini and co-workers,[22] in the above mentioned case study of intensive dairy farm situated in the Po valley (Italy): GHG emission reduction due to AD, calculated as equal to −23.7%, can reach −36.5% when a gas-tight tank is used for digestate storage.

A proper design and management of feedstock and digestate storage units looks also important in order to mitigate the odour impact of the plant. Indeed, the two major sources of the olfactory annoyance are biomass storage production of biogas and digestate composting units.[47] Closed-operated hydrothermal hydrolysis has positive effects on overall fugitive odour control in plants; on the other hand, eventual fugitive emissions during high-temperature and seemingly open pre-treatments can be the principal source of odours.[48]

In conclusion, gas tight storage should always be advised, since the corresponding GHG and ammonia fugitive emissions are even more important those coming from fertilizers.[49] As mentioned above, avoiding leakages and using closed tanks are among the most important ways to reduce the global warming impact of biogas plants.[19]

Impact of digestate final use

The use of agricultural and zootechnical byproducts and MSW as soil improver and fertilizer is a sustainable approach, allowing to reduce the production, transport and use of synthetic chemicals: however, spreading untreated biomass on soils sometimes implies the release into the atmosphere of huge amounts of chemicals such as methane, nitrous oxide, ammonia, volatile hydrocarbons, etc. Anaerobic digestion of biomass followed by the use of digestate as biofertilizer is a common practice related to biogas production. In this paragraph, the current knowledge concerning the environmental impact of this practice is briefly discussed.

A recent study on this topic[50] concluded that direct effects of anaerobic digestion on long-term sustainability in terms of soil fertility and environmental impact at the field level are of minor relevance; indeed, the most relevant issue (with regard to both emissions to atmosphere and in soil fertility) is related to possible changes in cropping systems. According to this study, the main direct aftermaths of anaerobic digestion are short-term effects on soil microbial activity and changes in the soil microbial community. Considering soil quality, digestate is significantly more inert vs. atmospheric and biological agents than the biomass itself: this property results into a lower degradation rate of the organic matter. In fact, labile fractions of original biomass such as carbohydrates are rapidly degraded, causing the enrichment of more persistent molecules such as lignin and non-hydrolysable lipids.[51] In a specific case study on pig slurry anaerobic digestion, a high biological stability of biomasses was achieved, with a Potential Dynamic Respiration Index (PDRI) close to 1,000 mg O_2 kg VS^{-1} h^{-1}.[10]

With regard to nitrate leaching and release into the atmosphere of ammonia and nitrous oxide, the current state of knowledges needs to be improved: however, the impact is considered "negligible or at least ambiguous".[50] The "ambiguity" of previous studies, as highlighted by this Author, is probably due to the different impact of digestate depending on the type of considered soil. For instance, Eickenscheidt and co-workers[52] investigated the emission of methane, nitrous oxide and ammonia from untreated manure and digestate applied on several soils: while methane emissions did not significantly change, high N_2O emissions were observed in the correspondence of high carbon loadings. A significative impact of soil moisture-soil mineral-N interactions on N_2O emissions was also observed by Senbayram and co-workers.[31]

Considering N_2O and CH_4, digestate can give rise to significant emission rates into the atmosphere: however, these emissions are generally lower than untreated biomass.[53] As for nitrous oxide, digested products are more recalcitrant than fresh slurry; thus, microbial degradation is slower, in which leads to relatively few anoxic microsites and poor N_2O emission compared to fresh slurry application.[54–56] Conversely, methane emissions from digestate are generally lower than those of original biomass, since the methanogenic potential is reduced: this is particularly relevant in the presence of reduced methane coming from manure[26,45] (Poeschl et al., 2012; Boulamanti et al., 2013). As for methane emission, an exception is known in the specific case of rice cultivation: indeed, adding digestate to paddy results into the methane emission rate enhancement from 16.9 to 29.9 g m^{-2},[57] whilst no significant effects are observed for N_2O.[57,58]

Based on the above-cited literature, N_2O and CH_4 emissions from digestate are not critical, while ammonia release and nitrate leaching are still a critical point. For instance, ammonia emissions from digestate higher than from original manure have been observed in several studies.[56,59,60] It was also reported that up to 30% of nitrogen can be lost by ammonia volatilization, due to the enhancement of soil pH.[59,60] Specifically, Matsunaka and co-workers[61] reported a 13% nitrogen volatilization as ammonia, when anaerobically digested cattle slurry was used as soil fertilizer for grassland. The practice of fertilizing soil with anaerobically digested materials increases soil concentration of NO_3^- (+30/40% compared to raw cattle slurry): this is associated to the four times more readily degradable organic C increased microbial biomass, depleting nitrogen and oxygen concentration in soil and resulting in the 10 times increase of CO_2 and N_2O emissions.[62] A proper management of digestate can mitigate its environmental impact: ammonia emission rates ranging from 1.6 to 30.4 were reported, depending on the adopted practice.[63]

With regards to pesticides, heavy metals and harmful microorganisms, the risk of food chain contamination is generally considered low,[64] but the soil burden of persistent organic pollutants (POPs) caused by the use of digestate as biofertilizer still needs to be fully assessed.[65] On the other hand, anaerobic digestion can have relevant effects on phytotoxicity of specific biomass: for instance, the phyto-toxic character of olive mill effluent is reduced after anaerobic digestion,[66] and the degradation of aflatoxin B1 from corn grain can be reached.[67] Finally, an odour reduction up to 82–88% can be obtained.[63]

In conclusion, the main critical issue in final use of digestate is nitrogen release into the environment, which can be reduced by applying the best practices for preserving soil quality. The management of nitrogen dosage is sometimes difficult because of the feedstock variability. It is also important to remark that fugitive emissions from digestate storage are generally more important than those released by its use into soil, as indicated above.[20,49]

Impact on particulate matter

With regards to particulate matter (PM), biogas combustion is not a significant emission source when compared to other fuels: emission factors of 0.238 and 0.232 g/Nm^3_{CH4} have been estimated for flaring and CHP, respectively.[34] However, secondary PM formation can occur, due to NO_x emissions from CHP and NH_3 volatilization from storage and digestate final use. Indeed, during secondary PM formation, the prominent roles of ammonia[68] and NO_x[69] are ascertained. As reported by Boulamanti and co-worker,[45] NO_x emissions are in general the principal source of secondary PM from biogas. As discussed above, closed storage can significantly abate ammonia emissions, resulting also into the global reduction of PM formation from this contaminant.

Impact of biogas upgrading to biomethane

Biomethane production is an efficient approach to increase the market share of biogas, resulting in a further reduction of fossil fuels. The equivalent CO_2 saving raises considerably if methane slip is limited to 0.05%,[70] while the process results no longer sustainable when methane losses reach 4%. Biomethane use as an alternative to gasoil is expected to improve local air quality, with regards to NO_x and particulate matter. As a consequence, biogas upgrading for vehicle fuelling purposes produces optimum benefits with respect to photochemical oxidant formation, marine eutrophication and ecotoxicity; on the other hand, scarce benefits are observed in terms of climate change compared to biogas combustion in CHP.[71]

Depending on several factors such as energy consumption, production and transport of materials used, produced waste and methane slip, the environmental impact of biomethane production depends on the upgrading technology adopted. In PSA, the eventual recovery of the off-gas plays a key role.[72,73] Starr and co-workers[74] reported that the most CO_2-efficient upgrading technology for MSW biogas is the BABIU (bottom ash upgrading) based on ash produced by municipal waste incinerators. The condition required is that the incinerator lies within 125 km from the biogas upgrading plant. Considering water scrubbing in basic solutions, a lower impact can be achieved by replacing KOH with NaOH. Water from biogas upgrading plants can be recycled in the process or treated as wastewater, depending on chemical composition: the most common VOC in the wastewater of biogas upgrading plants are p-cymene, d-limonene and 2-butanone[75]; the maximum VOC content is observed in MSW treatment plants, reaching up to 238 mg/L, but no inhibition is observed when wastewaters are recycled in the plant.

Along its impact on climate, biomethane use as gasoil substitute of is expected to improve urban air quality, because emission factors of methane are up to 10 times lower than those of liquid fuels, considering PM, VOCs and polycyclic aromatic hydrocarbons.[76] Biomethane injection in the national grid may also reduce residential solid fuels consumption in some specific regions, with relevant benefits on indoor air quality and human health.[77]

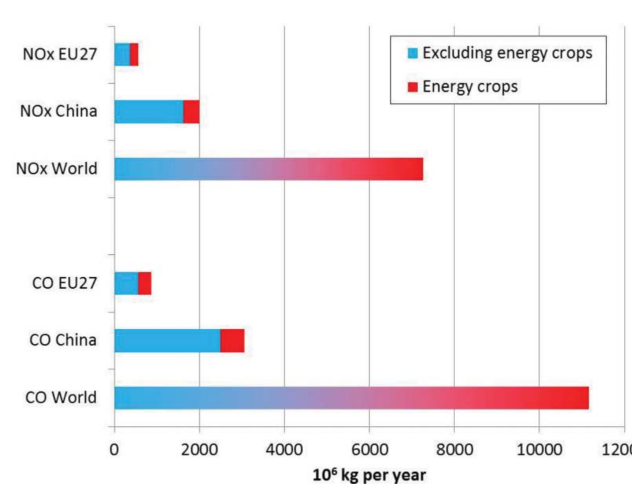

Figure 2. Emission potential of biogas plants for NO_x and CO.

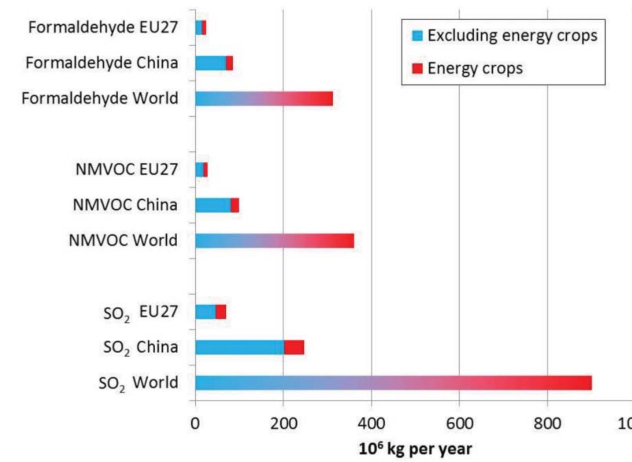

Figure 3. Emission potential of biogas plants for formaldehyde, NMVOC and SO_2.

Global emission potential

The potential emission associated to biogas plants is reported in Figure 2 (NO_x and CO) and in Figure 3 (for formaldehyde, NMVOC and SO_2). Data are obtained combining emission factors reported in Table 1[25] and energy potential reported in Figure 1. For Europe and China, the contribution of energy crops is reported separately, since their use is often disregarded due to its negative impact on land availability for food. In the case of the global potential, the relative contribution of energy crops is not available.

Conclusions

Biogas can significantly contribute to abate greenhouse gas emissions. However, attention must be payed towards undesired emissions of methane and nitrous oxide (N_2O). The emission budgets of the two compounds are scarcely related to direct release from biogas/biomethane combustion, whilst biomass storage and digestate management are the critical steps. Similar considerations apply to ammonia: to reduce its impact on secondary aerosol formation, efficient biomass and digestate storage should always be recommended. Among all the gaseous pollutants considered in direct emission from biogas combustion, nitrogen oxides (NO_x) level were worth of some concern in several case studies. On the other hand, volatile organic compounds do not seem to constitute a critical issue. Considering the aftermaths of digestate spreading on soil quality, further studies are needed in order to fully assess the long-term impact. In the medium-short term, digestate seems to be preferable compared to untreated biomass. The upgrading to biomethane can generally improve air quality and reduce GHG emissions; however methane losses in the off-gas can affect the sustainability of the whole process.

Acknowledgments

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