ANTHROPOGENIC EMISSIONS OF METHANE AND NITROUS OXIDE IN THE FEDERAL REPUBLIC OF GERMANY

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Abstract. The anthropogenic emission sources of methane (CH_4) and nitrous oxide (N_2O) in the Federal Republic of Germany were investigated. The object of the recently completed first phase of this research project was to summarize the present knowledge about the emission sources, make a first rough estimate of the emissions, identify the need for further research in the field, and - as far as possible - discuss the existing possibilities to reduce emissions. The main CH_4 emission sources identified are the landfills, stock farming and pit mining, the main N_2O sources are agriculture (including a minor contribution from animal wastes) and the production of adipic acid, the latter possibly being reducible by means of a new catalytic process. The total anthropogenic emissions of CH_4 from Germany are estimated roughly at 5.4 - 7.7 million tonnes per year, contributing a share of 2 % to the world-wide anthropogenic emissions. Those of N_2O are estimated roughly at 200 000 - 280 000 tonnes per year, contributing a share of 3 - 4 %.

1. Introduction

In summer 1993, the Fraunhofer-Institute for Systems and Innovation Research (FhG-ISI) finished the first phase of a research project which dealt with anthropogenic emissions of the climate relevant trace gases nitrous oxide (N_2O) and methane (CH_4) in the Federal Republic of Germany (Schön, Walz et. al., 1993). For the first time it was possible to calculate a representative emission data base for different emission sectors in Germany. However, due to the limited time and funds available for the first phase, the resulting emission quantities can only be viewed as temporary and rough estimates. The main results are presented in the following summary.

2. Emissions

2.1. METHANE

Table I gives an overview of CH_4 emissions in the Federal Republic of Germany. The largest emission sources are the landfills, stock farming and coal mining which together account for more than three-quarters of recorded total emissions.

TABLE I

Rough estimate of CH_4 emissions in the Federal Republic of Germany.

emission sector	CH_4 emission in kt/a
landfills *	1 800 - 3 150
- sewage sludge deposition	64 - 105
stock farming	1 900
- metabolism	1 400
- animal waste	500
coal mining	970 - 1 760
- hard coal	962 - 1 400
- lignite	8 - 360
gas and mineral oil	338
- gas old Federal States	266
- gas new Federal States	68
- mineral oil	4
waters	252 - 383
- surface water	9 - 25
- ground water	188
- wetlands	55 - 170
waste water treatment **	50 - 64
combustion of fossil fuels ***	60
transportation	35
industrial processes (only partially investigated)	negligible
biomass burning	4 - 7
total	5 409 - 7 697

* including sewage sludge deposition
** excluding sewage sludge deposition
*** preliminary results of Federal Environmental Agency

Temporally varying amounts of CH_4 are given off from depositing waste containing decomposable organic carbon compounds. The estimate of the CH_4 potential is still very uncertain. For example, it still has to be investigated to what extent emissions are influenced by the breakdown of carbon compounds which are only partly degradable, by carbon discharges from seepage water and by oxidation of CH_4 to CO_2 within the dump. The annual yield of CH_4 in landfills is estimated at approx. 2 340 - 4 090 kt. After subtracting the amount that is burnt off or used for energy, there remains an annual CH_4 emission of 1 800 - 3 150 kt. Further research should concentrate on estimating the gas potential and investigate ways of reducing emissions.

Stock farming makes a large contribution to CH_4 emissions. Metabolically caused emissions from livestock in Germany are about 1 400 kt/a, of which the predominant part can be traced to ruminants and cattle in particular. Since CH_4 emissions during fermentation of raw fibres in the rumen of ruminants are energetic losses, questions about these metabolic losses have already been intensively investigated in the past for economic reasons. So, this value can be judged as relatively certain.

CH_4 emissions from the anaerobic degradation of organic substances in animal waste have to be added to this. These vary a lot depending on the type and duration of storage as well as the treatment of the waste. There are no reliable quantitative data available for this sector in contrast to the emissions from metabolic sources. Only the CH_4 potential can be calculated with sufficient accuracy. The value given for this sector of approx. 500 kt/a can only convey an order of magnitude of the CH_4 actually emitted. In order to get a sound estimate it would be necessary to have information about the proportions of storage and treatment methods. In addition, measurements of emissions at known substrate compositions dependent on different types of storage of cattle and pig manure would be necessary.

CH_4 is also formed during the formation of coal from dead vegetation which is then released during the extraction and processing of the coal. In the pit mining sector, reliable data are available due to the safety precautions. Taking into account the burning and energetic utilisation of the gas, there are CH_4 emissions of 962 - 1 400 kt/a. The data available from lignite open-cast mining is much less reliable; emissions are estimated between 8 - 360 kt/a. Further research should corroborate data from open-cast mining and investigate the state of development of innovative reduction techniques such as the catalytic combustion of CH_4 or its concentration using molecular sieves.

The gas and oil industries, waters, sewage purification, combustion of fossil fuels, and the transport sector play a less significant role. Emissions from industrial processes and from biomass combustion can be judged as insignificant within the Federal Republic of Germany.

Finally, it should be pointed out that agricultural land use can also indirectly increase the concentration of CH_4 in the atmosphere as first research results indicate that nitrogen fertilizers reduce the capacity of the soil to act as a CH_4 sink.

Altogether, the anthropogenic emissions of CH_4 in Germany (with one major exception: waters and marshes, where it is hardly possible to distinguish between natural and anthropogenic sources) total approx. 5 400 - 7 700 kt/a or roughly calculated 2 % of the global anthropogenic emissions (350 million t/a).

Despite the limitations regarding the exact amount of emissions it is still possible to make initial statements about available reduction possiblities. There is a significant reduction potential in landfills and coal mines via improved energetic utilisation of pit gas and landfill gas respectively. The energetic utilisation of CH_4

from animal waste, for example in biogas reactors, would also result in a promising reduction of emissions. These measures would bring additional advantages due to the possible substitution of fossil fuels. High priority must also be given to renewing the gas grid in the new Federal States particularly since the switch to natural gas is currently in progress there. A significant proportion of the CH_4 emissions from sewage treatment plants in the new Federal States could be avoided if mesophilic sludge stabilisation taking place in closed containers would substitute for the presently common psychrophilic way.

2.2. NITROUS OXIDE

Rough estimates of the anthropogenically caused N_2O emissions in the Federal Republic of Germany are summarised in Table II. The industrial processes and the agricultural sector are the largest emitters with a share of approx. 37 - 40 % of the total emissions and 32 - 38 % respectively followed by the waters sector (12 - 23 %).

N_2O emissions from industrial processes have not been accounted for in any detail in previous literature. The main sources of N_2O identified are the manufacture of adipic acid and nitric acid, production and use of N_2O (e.g. for narcotics) and possibly large-scale conversions of nitric acid and other nitrogen compounds.

Assuming a specific emission factor of 333 kg N_2O/t adipic acid, the total emissions resulting from the process of adipic acid manufacture for the old and new Federal States are in the order of 78 - 86 kt/a. Thus, adipic acid production is one of the most significant anthropogenic sources of N_2O emissions in Germany. To reduce this considerable amount, it is planned to use catalystic processes, although the reduction in emissions that is hoped to be achieved cannot be given at present.

Calculations made on the basis of specifications in literature and from industry indicate that between 5.5 and 11 kt of N_2O are emitted annually due to nitric acid production. Assuming that the total quantities of N_2O used in narcotics and for other applications are also emitted during use, then the N_2O emissions from N_2O production and use can be estimated at approx. 5 kt per year.

In total, the annual emission of N_2O from the industrial processes examined and N_2O use is estimated at 83 - 102 kt.

The applied quantity of nitrogen fertilisers is decisive for the absolute amount of N_2O emissions from the soil. Specific emission factors of 0.9 - 4.3 kg N_2O per hectare and year have been calculated for fertilised areas, dependent on the type of soil, crop and varying climatic relationships during the individual sampling times. If specific emissions are assumed of 2 kg and 3 - 4 kg N_2O-N per hectare and year for cereal fields and grassland respectively, then N_2O emissions for the area of the Federal Republic total 60 - 70 kt/a.

TABLE II

Rough estimate of N_2O emissions in the Federal Republic of Germany.

emission sector	N_2O emission in kt/a
industrial processes	
(only partially investigated)	83 - 102
- N_2O production and use	0 - 5
- nitric acid production	5 - 11
- adipic acid production	78 - 86
agriculture and stock farming	78 - 88
- agriculture	60 - 70
- pasture	7
- agricultural waste *	11
waters **	24 - 64
- surface water	6 - 22
- ground water	18 - 42
combustion of fossil fuels	
(only large furnaces)	17
transportation	4
waste water treatment ***	0.4 - 0.5
composting ****	negligible
biomass burning	negligible
total	206 - 276

* indirect effect from NH_3 emissions
** including N_2O remaining in sewage
*** excluding N_2O remaining in sewage
**** excluding effect from NH_3 emissions

7 kt/a N_2O from nitrogen entry into the soil due to pasturing cattle must be added to this. In addition, the extraction and storage of agricultural waste and the corresponding ammonia emitted also causes an indirect nitrogen input to the soil. The resulting indirect N_2O emissions are estimated at 11 kt/a.

Accordingly, a first rough estimate for total N_2O emissions from the agricultural sector including pasture of cattle is 78 - 88 kt/a.

N_2O can also be formed during nitrification and denitrification processes in surface waters and groundwater acquifers mostly due to the increasing amount of nitrogen compounds of anthropogenic origins entering the waters. According to the specific emission factors given in the literature (mainly U.S. figures which still have to be validated), 6 - 22 kt/a of N_2O is yielded from surface waters which simultaneously includes the little quantity of N_2O remaining in sewage from purification plants. A N_2O production of 18 - 42 kt/a has been calculated on a simi-

lar basis for groundwater acquifers. Assuming that this is emitted completely into the atmosphere, a total amount of 24 - 64 kt/a N_2O is given off from waters.

In the past, errors in taking flue gas samples led to overestimation of emissions from furnaces. Nevertheless, the combustion of coal in particular is a relevant emission source, while the transportation sector and sewage plants play a minor role albeit with an increasing influence. This is due to the tendency, respectively to equip vehicles with catalysts, where at low operating temperatures N_2O may be formed and to equip the sewage purification plants with biological devices to eliminate nitrogen, where small amounts of N_2O may be formed as well. Composting and biomass combustion can be assessed as being relatively insignificant.

The rough estimate indicates the total amount of approx. 200 - 280 kt/a N_2O emitted from anthropogenic origins in Germany. This corresponds to a share of 3 - 4 % related to the worldwide anthropogenic N_2O emissions (6 - 9 million t/a).

3. Future Research Projects

A whole wealth of questions arose in practically all the areas of investigation. It will be the task of further research to set up sound emission accounts for every emission sector and to analyse existing and future emission reduction possiblities so that it will be possible to devise concrete strategies of action and show the research requirements that are left.

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References

Schön, M., Walz, R. et al.: 1993 (forthcoming), 'Emissionen der Treibhausgase Distickstoffoxid und Methan in Deutschland', Federal Environmental Agency (UBA; Ed.), *UBA report 9/93,* Erich Schmidt Verlag, Berlin.