Chemosphere, Vol.26, Nos.1-4, pp 447-452, 1993
Printed in Great Britain

0045-6535/93 $6.00 + 0.00
Pergamon Press Ltd.

# A Global Methane Emissions Program for Landfills, Coal Mines, and Natural Gas Systems

L.L. Beck

The U.S. Environmental Protection Agency
Office of Research and Development
Air and Energy Engineering Research Laboratory
Research Triangle Park, North Carolina 27711 USA

(Received in USA 17 October 1991; accepted 21 May 1992)

ABSTRACT

The Air and Energy Engineering Research Laboratory (AEERL) of EPA's Office of Research and Development has chosen anthropogenic methane emissions as a principal focus in its global climate research program. Three of the major sources are municipal solid waste landfills, coal mines, and natural gas systems. This paper presents the scope and methodology of the AEERL methane emission studies and discloses data accumulated thus far in the program. A major emphasis in the landfill program is measurement of emissions from operating landfills and calculation of country-specific emissions. Landfill methane emissions are not estimated, but factors affecting emissions are discussed and estimates developed by others are provided. For coal mines, existing data collected by other researchers on underground mines are combined with EPA data on emissions from surface mines to provide an estimate of global emissions of 43 Tg/yr. Methane from natural gas production, transmission, and distribution systems is estimated to be 4.4 Tg/yr for the United States.

## 1. INTRODUCTION

The U.S. Environmental Protection Agency's (EPA's) Office of Research and Development has been providing research since 1988 on the magnitude and sources of radiatively important trace gases (RITGs), with special emphasis on atmospheric methane ($CH_4$). $CH_4$ was chosen as an area of primary concern because of its short atmospheric lifetime (10 years) relative to some of the other RITGs such as carbon dioxide (50-200 years); chlorofluorocarbons (65 years for CFC-11, 130 years for CFC-12); and nitrous oxide (150 years) (IPCC, 1990). Because of its relatively short lifetime and the fact that the abundance of $CH_4$ increases is due to human activity, the EPA believes that efforts to stabilize atmospheric concentrations of this important RITG have the potential to produce positive results in a relatively short time frame (EPA, 1990a).

The Air and Energy Engineering Research Laboratory (AEERL) of EPA's Office of Research and Development is compiling data on $CH_4$ emissions from all sources. However, the AEERL has decided to focus initially on three major sources of $CH_4$: municipal solid waste (MSW) landfills, coal mines, and natural gas systems. These three sources comprise an estimated 20 percent of anthropogenic $CH_4$ globally (EPA, 1990a), and may be more amenable to cost effective mitigation measures than $CH_4$ emissions from other human-related activities such as rice cultivation, enteric fermentation, and biomass burning. Also, there is a strong possibility that mitigation measures can utilize the recovered $CH_4$ from these sources as fuel. This would reduce RITG emissions (e.g., carbon dioxide) which would otherwise be released from the burning of fossil fuels. Mitigation of these sources could therefore be justified for reasons apart from global warming concerns.

There is uncertainty in the magnitude of emissions from landfills, coal mines, and natural gas systems. First, there are different emission characteristics among specific categories of sources and individual sources within the categories. These differences are quantified by developing "emission factors" (quantity of emissions per source). Similarly, there is uncertainty in the numbers and kinds of sources within each country, or "activity data." The AEERL has chosen first to reduce the uncertainty in national and global emission inventories by developing more accurate emission factors and activity data (emission factor x activity data = inventory). An additional programmatic emphasis is to develop first inventories of U.S. sources, then global inventories. This is because of the accessibility of sources and the need in some cases to develop or refine methodology for measuring or estimating emissions.

Information on RITG emissions is being developed in cooperation with the global climate research community, and is being coordinated with international agencies such as the Organisation for Economic Co-operation and Development (OECD). The information which follows is accurate from a technical perspective; however, nothing in this paper should be construed to represent EPA policy.

## 2. EMISSIONS FROM LANDFILLS, COAL MINES, AND NATURAL GAS SYSTEMS

### A. Municipal Solid Waste (MSW) Landfills

$CH_4$ is generated from MSW landfills by the anaerobic decomposition of organic material. Current estimates of $CH_4$ emissions from MSW landfills range from 25 to 40 teragrams per year (Tg/yr) (EPA, 1990a) (a teragram is $10^{12}$ grams or 1 million metric tonnes), and emissions can continue for 100 years or longer (Thorneloe, 1991). The major variables being evaluated which are expected to influence the volume and rate of emissions are waste composition, temperature, moisture, disposal method, and time. The AEERL is developing emission factors for several types of landfills in a field testing program (Thorneloe and Peer, 1991). To date, research is in progress at AEERL to collect data at more than 30 landfills to develop more reliable emission estimates. Research is also being initiated to correlate $CH_4$ emissions with biodegradable components in landfills (i.e., food vs paper vs yard waste) and to validate a procedure which uses shallow probes to determine gas emission potential directly from specific landfills.

Another important factor in determining global landfill-$CH_4$ emissions is the total amount of refuse in place. This total for any given country in a given year is the sum of the MSW landfilled that year plus the landfilled MSW remaining from previous years. Country-specific data being collected by AEERL on the amount and composition of landfill waste will provide data to correlate $CH_4$ generation rate with key variables.

Climate for a given country is also an important variable since $CH_4$ generation rates are dependant on moisture, temperature, and pH (Thorneloe and Peer, 1990). Because of this, the AEERL program will consider dividing large countries such as the U.S. and Canada into climatic regions.

Waste composition is another important consideration when estimating the $CH_4$ generation potential for a given landfill, and an AEERL report on the effect of waste composition on landfill air emissions is planned for June 1992 (Thorneloe, 1991). As might be expected, the greater the proportion of paper, wood, textiles, and other biodegradable materials, the higher the $CH_4$ emission potential. Data are available on the waste composition for industrialized countries such as the U.S., the U.K., and Canada. Also waste composition is available for some of the major developing countries such as India. The AEERL will develop estimates where data are not available.

Finally, the use of systems to collect and utilize or dispose of (e.g., flare) landfill gas will affect the potential for a landfill to emit $CH_4$ to the atmosphere. The U.S. has the greatest number of existing and planned landfill collection projects, followed by Germany, the U.K., and Sweden (Thomeloe and Peer, 1991).

As this brief discussion indicates, landfill emission rates depend on the interrelationship of many complex variables, most of which have a non-linear relationship to $CH_4$ generation. The AEERL has developed a landfill $CH_4$ emissions model to assist in processing these data to determine $CH_4$ emission rates from MSW landfills (Peer and Epperson, 1992). The AEERL is also continuing to refine the input data and algorithms. In addition to the physical parameters which appear to affect chemical reaction kinetics, $CH_4$ emissions from MSW landfills appear to correlate well with some demographic data such as population and gross national product (GNP) (Peer, et al., 1991).

The AEERL is also conducting a demonstration for operating a 200 kW, commercially available, phosphoric acid fuel cell on landfill gas. Using this technique, the first of its kind, landfill $CH_4$ is reduced and electricity is produced as a byproduct. Fuel cells are a potentially superior technology to boilers, internal combustion engines, and gas turbines for mitigation of landfill $CH_4$ because they are highly efficient, quiet, and appear to be environmentally clean. This 4-year project will include a 1-year demonstration of the fuel cell and landfill gas cleanup equipment.

B. Coal Mines

$CH_4$ is trapped in coal seams and is released to the atmosphere when the coal seam is exposed to the atmosphere. This happens during mining operations and also during pre-mine degassing operations performed at some mines. The amount of $CH_4$ in the seam is a function of coal age, moisture, and depth. The amount and rate of release to the atmosphere depends on the physical and chemical characteristics of the coal, mining techniques, and degassing (if performed). $CH_4$ is also released from abandoned mines and to a small extent by natural processes such as erosion and diffusion from the coal seam to the Earth's surface.

Research into coalbed $CH_4$ generation has historically been performed primarily for safety reasons. This has been to protect miners because of the explosive nature of $CH_4$ when mixed with air in concentrations ranging from 5 to 15 percent. Because of this, the practice has been to evacuate the $CH_4$ from the mine or to dilute it to less than 1 percent prior to entry by the miners. This has been accomplished through mine ventilation and more recently by gob gas wells and pre-mine degassing.

Mines are most commonly ventilated by using continuously operating fans which circulate fresh air across the actively mined coal face. Because of the large volumes of air needed to keep $CH_4$ concentrations below the lower explosive limit, ventilation air contains less than 1 percent $CH_4$ and therefore has little or no economic value. It is vented directly to the atmosphere.

$CH_4$ is also released from gob wells. Underground mines can release large amounts of $CH_4$ when the fractured area behind the working longwall face collapses. This collapsed stratum, called "gob," releases $CH_4$. To prevent this $CH_4$ from entering the mining area, gob wells are drilled 2 to 15 m above the area being mined prior to the mining of the longwall panel (EPA, 1990a). The well operates at a negative pressure and thus removes mine air. Consequently, most gob gas is usually vented to the atmosphere. There are exceptions, however. In Alabama over 849,000 $m^3$/ day from 80 gob wells is captured and sold as natural gas. Since the program was initiated, over $1.1 \times 10^9$ $m^3$ of $CH_4$ has been used as fuel rather than vented to the atmosphere (Boyer, et al., 1990).

Pre-mine degassing can produce fuel-quality $CH_4$ and is an excellent technique for reducing $CH_4$ emissions to the atmosphere. This is practiced for purely economic reasons at some locations with gassy coal, such as in the Black Warrior and San Juan Basins in Alabama and Colorado, respectively (The Energy Daily, 1989; GRI, 1990).

The AEERL evaluated existing estimates of $CH_4$ from coal mining and found that most estimates neglected one or more important variables. Most notably, most estimates do not consider emissions from abandoned mines or from surface mines, even though surface mines are responsible for approximately 40 percent of global coal production. Emissions from surface mines are less than from underground mines because these coal reserves are closer to the surface (may have "leaked" more), are younger, and have been exposed to less heat and pressure than the deeper reserves. During the geologic past, the deeper coals were covered by 10 km or more of overlying strata, resulting in high pressures and temperatures in excess of 300°C (Boyer, et al., 1990). $CH_4$ is a product of geochemical coalification, and heat and pressure are the two primary agents which drive the chemical reactions that comprise the coalification process.

Because of the paucity of data on $CH_4$ emissions from surface mines, the AEERL is undertaking a program to measure emissions from active surface mines. The methodology chosen is to make in-situ, open-path infrared spectroscopy measurements using a Fourier Transform Infrared Remote (FTIR) sensor. This technique involves directing an infrared beam across an open area to a reflector. The reflected beam is then subjected to absorption analysis to quantify the gases of interest along the path of the beam. The behavioral characteristics of a plume at a given site are approximated using a tracer gas (sulfur hexafluoride) and application of standard Gaussian dispersion equations (Kirchgessner et al., 1992a).

The AEERL measured emissions from the Caballo mine in Campbell County, Wyoming, which is the Wyoming Powder River region. The Powder River region is recognized for its coalbed $CH_4$ resources. Even so, the estimated $CH_4$ emission rate (based on measurements) of 62 million ft $^3$/yr was five times higher than would have been expected if only coalbed $CH_4$ content and coal production rate had been used.

Using emission measurements from surface mines, combined with data from underground mines collected by EPA and others, The AEERL has revised our global emissions inventory for $CH_4$ emissions from coal mines. The initial estimate based on published data and emission measurements is 36 Tg/yr for underground mines and 6.9 Tg/yr for surface mines for a total global estimate of about 43 Tg/yr (Kirchgessner et al., 1992b).

### C. Natural Gas Systems

For the purpose of the AEERL study, EPA defines "natural gas systems" as natural gas production, transmission, and distribution systems. This breakdown of the industry has been used by other researchers and covers the industry from the gas production wellhead to the exit of the end-use meter. Oil wells which produce natural gas are also included in the study. Quantifying leakage (emissions) from natural gas systems is important for several reasons. The leaks are significant from their direct injection into the atmosphere because they are predominantly $CH_4$. It is also important to know the leakage rate in order to evaluate the effectiveness of fuel-switching strategies. Several researchers have suggested switching from coal and other fossil fuels to natural gas as a short-term mitigation strategy until cleaner energy sources can be developed and implemented, since natural gas has 55-60 percent of the carbon per unit of energy as coal (EPA, 1990a; EPA, 1990b). In order for such strategies to be effective, however, the natural gas leakage rate must be known and subtracted from the combustion gains. Since the relative radiative forcing of $CH_4$ is 21 times that of carbon dioxide on a molecule for molecule basis and 58 times more gram for gram (IPCC, 1990), leakage in the systems carrying natural gas to the fuel-switched combustor could offset the combustion gains.

The AEERL has entered into a multi-year program with the Gas Research Institute (GRI) to quantify leakage rates for natural gas systems in the United States. As part of this program a spreadsheet has been developed to assist in tabulating what is known or estimated regarding leak rates. By using this methodology and attendant statistical evaluation, the initial estimate for natural gas system leakage is 4.4 Tg/yr for U.S. sources.

The AEERL/GRI study of $CH_4$ emissions from natural gas systems began in 1990 and is expected to continue at least through 1993. The study is currently evaluating data needs and methodology for filling data gaps. Some of the methodology being considered is:

- Release of tracer gas coupled with upwind/downwind sampling,

- Meter balance techniques,

- Use of "sniffing" devices like organic vapor analyzers to determine rates of leakage from valves and fittings, and

- Direct measurement of emissions by bagging the component and sampling the collected emissions.

Once emissions data are collected, statistical methodology will be used to apply the data to estimate U.S. emissions.

## 3. SUMMARY AND COMMENT

The AEERL has concurrent programs directed at reducing the uncertainty of estimates of $CH_4$ emissions from landfills, coal mines, and natural gas systems. These programs were begun in 1990 and are in various stages of completion. The near-term focus of the MSW landfill program is to develop emission factors from U.S. landfills in a field test program. The coal mine program is applying state-of-the-art, in-situ measurement techniques to establish $CH_4$ emissions from surface mines, and will combine these data with knowledge about underground mine emissions to estimate global emissions. The natural gas study is concentrating first on reducing the uncertainty of emissions estimates from U.S. sources, and will apply this knowledge to establish $CH_4$ emission estimates for other countries which have natural gas systems. Table 1 presents current estimates of emissions from MSW landfills, coal mines, and natural gas systems.

Table 1. Estimates of $CH_4$ Emissions from MSW Landfills, Coal Mines, and Natural Gas Systems (Tg/Yr)

| | Estimate | Range |
|---|---|---|
| MSW Landfills (Global) | N/A[a] | 20 - 70[b] |
| Coal Mines (Global) | 43 | 19 - 50[b] |
| Natural Gas Systems (U.S. Only) | 4.4 | 0.51 - 8.4 |

[a] N/A - Not Available

[b] Range given in IPCC (1990)

The AEERL is also researching $CH_4$ emissions from cookstoves and biomass burning, though these programs are not as mature as the areas highlighted by this paper. The AEERL is also planning to begin research on other anthropogenic sources of $CH_4$ emissions. Some of the areas being considered are industrial and hazardous waste landfills, waste water treatment lagoons, septic systems, disposal of agricultural wastes, open burning of municipal solid waste, and sludge disposal.

The information being collected on $CH_4$ and other RITGs will be entered into a computerized database management system being developed by AEERL. The software, called Global Emissions Data (GloED) system, will serve as a repository for data and will be updated as new or better data become available. The software will be available early in 1992 for peer review and appraisal by other researchers.

## REFERENCES

Boyer, C.M., J.R. Kelafant, V.A. Kuuskraa, et al. (1990). Methane Emissions from Coal Mining: Issues and Opportunities for Reduction, Office of Air and Radiation. EPA/400/9-90/008. September.

EPA (U.S. Environmental Protection Agency) (1990a), Methane Emissions and Opportunities for Control, Workshop Results of Intergovernmental Panel on Climate Change. EPA/400/9-90/007.

EPA (U.S. Environmental Protection Agency) (1990b), Policy Options for Stabilizing Global Climate, Office of Policy, Planning, and Evaluation. EPA Document No. 21P-2003.

GRI (1990), Quarterly Review of Methane from Coal Seams Technology, March, (Gas Research Institute, Chicago, IL). Vol. 7, No. 3. ISSN 8756-9655.

IPCC (1990), (Intergovernmental Panel on Climate Change), Climate Change The IPCC Scientific Assessment, Haughton, J.T., G.J. Jenkins, and J.J. Ephraums, Editors, (University Press, Cambridge). ISBN 0-521-40720-6.

Kirchgessner, D.A., S.D. Piccot, and A. Chadha (1992a), Estimation of Methane Emissions from a Surface Coal Mine Using Open-Path FTIR Spectroscopy and Modeling Techniques, Accepted for Publication in Chemosphere.

Kirchgessner, D.A., S.D. Piccot, and J.D. Winkler (1992b), Estimate of Global Methane Emissions from Coal Mines, Accepted for Publication in Chemosphere.

Peer, R.L., A.E. Leininger, B.B. Emmel, et al. (U.S. Environmental Protection Agency) (1991), Approach for Estimating Global Landfill Methane Emissions, Air and Energy Engineering Research Laboratory. EPA-600/7-91-002 (NTIS PB91-149534), January.

Peer, R.L., and D.L. Epperson (1992), Development of an Empirical Model for Methane Emissions from Landfills, Air and Energy Engineering Research Laboratory. EPA-600/R-92-037 (NTIS PB92-152875), March.

The Energy Daily (1989), How "Moonbeam Gas" Made Its Way from Dream to Market, July 18.

Thorneloe, S.A. (1991), U.S. EPA's Global Climate Change Program - Landfill Emissions and Mitigation Research, Paper presented at Sardina 91 Third International Landfill Symposium in Cagliari, Italy.

Thorneloe, S.A., and R.L. Peer (1991), EPA's Global Climate Change Program - Global Landfill Methane, AWMA Annual Meeting, Vancouver, B.C.

Thorneloe, S.A., and R.L. Peer, (1990), Landfill Gas and the Greenhouse Effect, Presented at the International Conference on Landfill Gas: Energy and Environment, Bournemouth, England.