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NPL Report DQM 98

THE UK METHANE EMISSIONS INVENTORY: A SCOPING STUDY ON THE USE OF AMBIENT MEASUREMENTS TO REDUCE UNCERTAINTIES

(Contract PECD 7/12/148)

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ISSN 0309-3050

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Approved on behalf of Chief Executive, NPL, by
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Executive Summary

We present a critical review of the UK methane emissions inventory, identifying new data that has allowed us to calculate revised estimates for some sources. We have also undertaken a detailed and explicit analysis of the uncertainties associated with each parameter, allowing us to identify those areas which are most in need of improvement.

Our best estimate for total UK emissions is 5.0 Mt/a with upper and lower bounds of 7.2 and 3.6 Mt/a. The greatest uncertainty in this estimate relates to emissions from landfill sites. We have estimated landfill emissions in such a way that the sources of uncertainty are highlighted and quantified. Our best estimate for these emissions is 1.9 Mt/a, with upper and lower bounds of 4.2 and 0.6 Mt/a. This new result is comparable to existing estimates in both magnitude and uncertainty.

We believe that the uncertainty in the estimate of landfill emissions could be reduced significantly over the next few years. An initial focussed programme of direct emission measurements on major sites should provide a reduction of around 25%. This programme would be concentrated on large sites of modern design which will be responsible for an increasing proportion of future landfill emissions. Hence, this programme together with other activities in the field, will lead to a better understanding of the remaining uncertainties and so lead to further improvements in the total uncertainty. Similar programmes could reduce the uncertainty in the estimated emissions from other sectors, but their impact on the UK total would be significantly less.

We have identified five activities or processes as anthropogenic sources not at present included in the emissions estimates. We have proposed a modest programme to evaluate these emissions, though their effect on the total UK methane inventory is likely to be small.

Consideration has also been given to the use of ambient measurements to verify emissions estimates on a regional or national scale. Such techniques would provide added confidence in the completeness and accuracy of the inventory, but are essentially untested at present.

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THE UK METHANE EMISSIONS INVENTORY:A SCOPING STUDY ON THE USE OF AMBIENT MEASUREMENTS TO REDUCE UNCERTAINTIES

Background

This report was prepared by the National Physical Laboratory (NPL), Warren Spring Laboratory (WSL) and AEA Technology under contract (No 7/12/148) from the Global Atmosphere Division of the Department of the Environment (DoE).

Introduction

The UN Convention on Climate Change signed in Rio de Janeiro in 1992 will commit participating countries to limit anthropogenic emissions and to protect sinks of the gases responsible for global warming. Part of this activity will include the publication and maintenance of inventories of anthropogenic emissions of greenhouse gases together with estimates of future emissions. These inventories will eventually be used to establish the baseline for future reductions. Consequently, under or over estimation of the emission rates of some gases could limit the effectiveness of future reduction strategies.

The UK already has a National Atmospheric Emissions Inventory (NAEI), produced by WSL under contract to the DoE (Air Quality Division), but it is known that there are uncertainties in the NAEI for various gases, including methane, which is the second most important in terms of its global warming potential. The status of the emissions inventory for methane has been studied by the a working group of the "Watt Committee on Energy" (on behalf of the Department of Trade and Industry). As a result of the work of the Watt Committee, and fresh examination of their emissions by various industries, the UK Inventory has a very different aspect to that which it had a few years ago. In particular, emissions of methane from landfill sites are now believed to account for between 13 and 83% of total UK methane emissions (with a best estimate of 39%).

At present, few of the estimates used in the methane emissions inventory are based on direct measurements of actual emission rates, this is primarily because techniques available to perform such measurements have not been available. In recent years a number of organisations in the UK have developed new measurement techniques that are capable of measuring methane emission rates directly. However, as a consequence of the extent and variability of methane sources in the UK, the task of making measurements that are representative of emissions across entire sectors is a complex one. It is therefore important to consider how measurement resources can be deployed cost effectively in order to be representative and to achieve the greatest improvement in the emission inventory.

This report evaluates the present status of the UK emissions estimate for methane, and

assesses the practical benefits that could be realised by applying available measurement techniques to the most important emission sources. The report is in three parts. The first is a thorough re-working of the UK National Atmospheric Emissions Inventory for methane, including, for the first time, estimates of the uncertainties associated with each parameter used. The second part contains a review of techniques available for the measurement of atmospheric methane emissions, together with an evaluation of the benefits of applying them to the task of improving the estimate of specific contributions to the total estimate of UK methane emissions. The use of these techniques to verify the total value and to investigate the possibility of omissions from the inventory is also discussed. The report concludes with recommendations for the most cost-effective methods for improving the inventory.

Part I Emissions of Methane from Anthropogenic Sources: Uncertainties in Present Estimates

1 INTRODUCTION

Estimates of methane emissions exist for sources such as landfill sites, coal mines and gas pipelines. In most cases, the emissions from these sources are not well quantified and large uncertainties exist in the estimates. Actual measurements of methane emissions have only been carried out on a limited scale and often the basis of estimates is not clear. Further sources of methane may also have been overlooked completely.

An accurate inventory of methane emissions is very important. The Government White Paper "This Common Inheritance" states that changes already under way in agriculture and energy recovery from landfill waste should reduce Britain's emissions of methane substantially in the next 30 years.' A good baseline inventory is necessary if progress towards reducing methane emissions is to be measured.

This report examines the emission estimates used in the National Atmospheric Emissions Inventory (NAEI) and those produced by the Watt Committee on Energy (1993). New data has been added where available. The uncertainty in these estimates has been represented in two ways: quantitatively, by calculating emission estimates as a range with a minimum and maximum estimate using risk analysis; and qualitatively by linking each emission estimate with a data quality rating. Each emission source is subdivided where possible and previously uncharacterised sources have been identified.

2 METHODOLOGY

2.1 General approach to estimating emissions

Emissions are in general estimated by combining an emission factor with a production statistic. An emission factor may be defined as the emission per process unit, for example, per tonne of product manufactured or per tonne of waste treated. The process unit should be easily quantifiable. The production statistic is the total number of process units. Emission factors may be derived from measurement of emissions, or they can be estimated based on knowledge of a process. Process statistics are usually taken from annual collections of statistics such as those produced by MAFF, DTI and the Central Statistical Office.

In many existing inventories a single value for emission factors and production statistics is fixed upon, giving a single value for the overall emission. Usually however, there is a great deal of uncertainty in both emission factor and production statistic. In this report, two techniques have been used to represent these uncertainties - risk analysis and data quality.

2.2 Use of risk analysis

Risk analysis has been used to calculate the distribution of emissions estimates from each source based on estimates of the distribution of each of the factors. This method involves the use of probability distributions rather than single values for parameters in calculations. Thus the uncertainty inherent in the emission factors and production statistics used to estimate emissions can be incorporated. Three types of probability distributions have been used in the calculations. These are normal, uniform and triangular distributions.

Normal - a normal distribution with a specified mean and standard deviation. In the following sections **N**(50,2) indicates a normal distribution with a mean value of 50 and a standard deviation of 2. For a normal distribution, approximately 95% of values will fall within 2 standard deviations of the mean and practically all values (99.7% will fall within three standard deviations). Normal distributions are useful to describe data sets which are derived from a large number of smaller data sets.

Triangular - triangular distribution with three points, lower bound, best estimate and upper bound. In the following sections **T**(10,20,50) indicates a triangular distribution with a minimum value of 10, a maximum of 50 and a most likely value of 20. A triangular distribution is useful as an approximate model of uncertainty when actual data is not known.

Uniform - a uniform rectangular distribution between a minimum and a maximum value. In the following sections **U**(10,20) indicates a uniform distribution between the values 10 and 20. A uniform distribution is useful where only an approximate range of values is known.

The calculation is performed using a Latin Hypercube method (similar to Monte Carlo) in which a value for each variable is picked randomly from within the probability distributions which have been set for that variable. The outputs are calculated, and the process is repeated for a set number of iterations with the outputs accumulated as a probability distribution. Thus by entering emission factors and production statistics as probability distributions the probable range of emissions can be calculated. The inputs for the risk analysis calculations are described in sections 3 to 13. The authors are not aware of previous work in the field using this technique.

2.3 Use of a data quality rating scale

As well as calculating the likely range of emissions for each source, a data quality has been assigned. This is a measure of both the quantity and quality of data used to calculate emissions and has long been used in the field by, for example, the USEPA (1985). Data quality for example reflects whether emission estimates are based on measurements or whether they are based on calculations. The more untested assumptions which are made, the lower the data quality will be.

The definitions given below are used generally when assigning data qualities to the estimates although a degree of subjectivity is unavoidable in the choice of rating.

A - An estimate based on a large number of measurements made at a large number of sites fully representative of the sector.

B - An estimate based on a large number of measurements made at a large number of sites representative of a large part of the sector.

C - An estimate based on a number of measurements made at a small number of representative sites or a calculation based on a number of relevant facts.

D - An estimate based on a single measurement or a calculation derived from a number of relevant facts and assumptions.

E - An estimate based on a calculation derived only from assumptions.

When estimates are produced by multiplying factors, the following combinations are used.

Inputs	Output
E * E	E
E * D	D
E * C	D
D * D	D
E * B	D
E * A	C
D * C	C
D * B	C

Inputs	Output
C * C	C
D * A	C
C * B	B
C * A	B
B * B	B
B * A	A
A * A	A

3 LANDFILL

3.1 Description of model

The amount of methane emitted from landfills has been estimated using a model which was originally developed by Warren Spring Laboratory (WSL) to compare methane and carbon dioxide emissions resulting from a number of waste management options (various combinations of amounts landfilled and incinerated). The model is based on weight arisings of waste types, data on the percentage of each waste falling into a number of categories and the degradable properties of each category.

The basic form of the model is;

$$P = \sum (M_n \times \sum (Comp_i \times CM_i \times CC_i)$$

and

$$E = P - \quad G_{ox} + G_{fl} + G_{util})$$

where

P = Theoretical production of methane in landfills
M_n = Mass of waste type n landfilled
$Comp_i$ = Proportion of category i in the waste
CM_i = Carbonaceous matter content of category i
CC_i = carbon content of carbonaceous matter of category i

E = Emission of methane
G_{ox} = methane lost by oxidation
G_{fl} = methane destroyed by flaring
G_{util} = methane utilised for energy

The carbon loss on degradation for each category is calculated from the carbonaceous matter and carbon content, and the amount of methane produced is determined by assuming that 50% of the degraded carbon is converted to methane (and 50% to carbon dioxide). The total amount of methane generated by each waste type is calculated by summing the amounts contributed by each of the categories. The total methane generated by landfill is calculated by combining the estimates for each waste type. A percentage of this production is deducted to account for oxidation of methane in the capping material, and the amounts flared and extracted for energy recovery are deducted to determine the actual emission. No assumptions are made with respect to the time period taken for wastes to biodegrade to levels achieved in an anaerobic digester - the annual production refers to 'steady state' conditions, ie the amount of waste is the same each year. The following sections provide information on the data used for each factor in the equations.

3.1.1 Mass landfilled (M_n)

The mass of different waste types landfilled has been determined from data on weight arisings and the percentage of each waste type which is landfilled. The basic data set for weight arisings has been published (Department of the Environment 1992). The categories used by the DoE are quite general, and for this study some subdivision has been made. The data used in the model is summarised in Table 1.

Table 1. Waste arisings and mass landfilled.

Waste Type	Annual arising (Mt)	Percentage disposed to landfill	Mass disposed to landfill (Mt/a)
Agricultural	80	0	0
Mining and quarrying			
- colliery and slate	51	10	5
- china clay	27	10	3
- quarrying	30	10	3
Sewage sludge	5	20	1
Dredged spoils	43	0	0
Household	15	85	13
Civic amenity	4	90	4
Commercial	15	100	15
Construction and demolition	24	55	13
Asphalt planings	8	20	2
Industrial			
- blast furnace and steel slag	6	30	2
- power station ash	13	50	7
- general	10	90	9
- misc processes	17	90	15
- food solids	3	80	2
- food effluent	15	100	15
- abattoir waste	2	70	1
- other	2	85	2
TOTAL	370	30	111

The main area of uncertainty in mass arisings is the amount of industrial wastes. DoE give arisings for blast furnace and steel slag and power station ash but group all remaining industrial wastes together. WSL have estimated that this consists of general wastes, similar to commercial waste (office and canteen waste from factories), residues and sludges from non-food production processes and food wastes (including solid wastes from food production (Gascoigne 1993), abattoir waste and sludges from the treatment of approximately 400 Mt/a of effluent produced by the industry). Other industrial wastes include hazardous wastes, clinical wastes, tyres and wood wastes.

Table 2 Inputs to risk analysis: waste arisings and mass landfilled

Source	Annual arising (Mt)	Percentage disposed to landfill
Agricultural	U(72,88)	0
Mining and quarrying		
- colliery and slate	U(46,56)	U(5,15)
- china clay	U(24,30)	U(5,15)
- quarrying	U(27,33)	U(5,15)
Sewage sludge	U(4.3,5.3)	U(15,25)
Dredged spoils	U(39,47)	U(0,10)
Household	U(13,16)	U(80,90)
Civic amenity	U(4,5)	U(85,95)
Commercial	U(12,18)	U(95,100)
Construction and demolition	U(22,26)	U(55,65)
Asphalt planings	U(7,9)	U(15,25)
Industrial		
- blast furnace and steel slag	U(5,7)	U(25,35)
- power station ash	U(12,14)	U(45,55)
- general	U(8,12)	U(85,95)
- misc. processes	U(14,20)	U(85,95)
- food solids	T(1.5,3,8)	U(75,85)
- food effluents	U(12,18)	U(90,100)
- abattoir wastes	U(1.4,2.1)	U(65,75)
- other	U(1.5,2.5)	U(80,90)

Data on the percentages disposed to landfills are partly taken from published sources and partly from estimates made by the authors. The uncertainties in both arisings and in percentages landfilled are in some cases considerable and therefore ranges have been used for all of the data. Table 2 summarises the ranges and distribution types used.

The largest uncertainties are those for commercial and industrial wastes, especially those from the food industry.

3.1.2 Composition of waste types ($Comp_i$)

It is assumed that five basic categories can be used to represent any waste arising in order to calculate the theoretical methane generated from landfill. The five categories are non-degradable organics (NDO), poorly degradable organics (PDO), readily degradable organics (RDO), inerts and 'free' water. The typical components contained in each category are as follows:

Table 3 Typical components of waste categories

Category	Typical components
Non degradable organics	Oil based synthetics
Poorly degradable organics	Cellulosics
Readily degradable organics	Putrescibles
Inerts	Minerals and metals
Free water	Water from sludges

Each waste type (household, commercial, industrial etc) is assumed to contain some or all of these categories and their fractions have been estimated. For arisings where good analytical data are available, such as household dustbin waste, it is possible to use precise values, but for other waste arisings it has been necessary to use ranges and 'best estimates' which are based on WSL knowledge of the waste type. Table 4 shows the values used in the model for the three categories of organic matter. The wastes may also contain inert matter and free water although these are not shown.

Table 4 Composition of wastes

Waste type	Percentage in category		
	NDO	PDO	RDO
Agriculture	-	-	-
Mining and quarrying			
- colliery and slate	0	0	0
- china clay	0	0	0
- quarrying	0	0	0
Sewage sludge	3	20	32
Dredged spoils	0	5	0
Household	15	35	25
Civic amenity	T(7,13,17)	T(15,22,30)	T(30,45,60)
Commercial	T(10,15,20)	T(30,60,80)	T(10,15,20)
Construction and demolition	5	5	0
Asphalt planings	T(0,5,10)	0	0
Industrial			
- blast furnace and steel slag	0	0	0
- power station ash	0	0	0
- general	T(15,20,25)	T(30,40,60)	T(10,15,20)
- misc. processes	U(0,10)	U(0,10)	0
- food solids	0	10	T(70,80,85)
- food effluent	0	5	U(45,55)
- abattoir wastes	0	10	T(70,80,85)
- other	35	35	15

3.1.3 Carbonaceous matter and carbon content (CM_i & CC_i)

For each category (NDO, RDO, etc) properties have been assigned in terms of carbonaceous matter (loss on degradation) and carbon content of the carbonaceous matter. Ranges and best estimates for these properties were obtained from the literature and from experimental data and these are shown in Table 5.

Table 5 Properties of categories used in the model

Category	Carbonaceous matter (loss on degradation) Wt %	Carbon content of carbonaceous matter Wt %
Non degradable organics	T(0,4,10)	T(40,80,90)
Poorly degradable organics	T(10,30,50)	T(30,40,50)
Readily degradable organics	T(10,20,40)	T(30,40,50)
Inerts	T(0,0.5,1)	T(30,50,90)
Free water	0	0

3.1.4 Losses due to oxidation (G_{ox})

Research indicates that methane oxidation (to $CO_2 + H_2O$) in surface capping may occur which could reduce levels escaping to atmosphere. Values of less than 5% to over 95% methane oxidation have been measured at individual landfill sites. For the model, the overall methane losses due to oxidation are estimated to lie between 10 and 40% of methane generation with a best estimate of 20% (ie a range of T(10,20,40)).

3.1.5 Flaring and utilisation of landfill gas (G_{fl} and G_{util})

Flaring is estimated at 360,000 tonnes per year. In January 1993 there were 51 landfill gas energy recovery schemes operating in the UK, with a total energy consumption of 300,000 tonnes of coal equivalent per year. This is approximately 150,000 tonnes/year of methane. Both flaring and utilisation will not be completely efficient at destroying methane, however losses from flares and other burners are likely to be small in comparison with direct emissions from the site.

3.2 Estimated emissions

The model's best estimate for the amount of methane actually emitted from landfills is 1.9

million tonnes of methane per year. The range of the model's estimates is from 0.6 to 4.2 million tonnes per year as shown in Figure 1.

The best estimate is comparable to the estimate of 2 million tonnes per year made by ETSU (Aumonier 1993). Their study of methane emissions from landfill estimated a similar range of 0.6 to 5.0 million tonnes per year. Despite this close agreement there are differences between the two calculations since the WSL model uses a different approach to that used by ETSU, and there are differences in much of the data used by the two studies. In particular, the models differ in the following ways:

- The present (WSL) calculation is strictly "steady state", while the ETSU calculation uses a 1st-order decay model and historic waste arisings data,
- Arisings of industrial waste are larger in the WSL calculation,

  Disposals to landfill of household, commercial and industrial wastes are different,
- Degradable carbon content of wastes varies between the models
- Level of methane oxidation assumed by the ETSU study is given greater level of uncertainty.

Differences between studies over this type of data is not surprising, and merely reflects the large levels of uncertainty in much of the available data.

In order that emissions can be predicted with greater accuracy by the WSL model (and the ETSU model) the uncertainties in much of the data need to be reduced, particularly:

- The composition of waste types, particularly: general industrial wastes, commercial wastes and food industry wastes

  the percentage of each waste disposed to landfill
- the carbonaceous matter content of the categories used in the model, especially the poorly degradable and readily degradable categories.
- the carbon content of the carbonaceous matter in each category.
- The losses due to oxidation in the surface capping.

In view of the large number of untested assumptions which have to be made in order to derive methane emission estimates from landfills, the estimate is given a data quality of D. Better data for both waste arisings and for methane generation is required before this data quality rating could be improved.

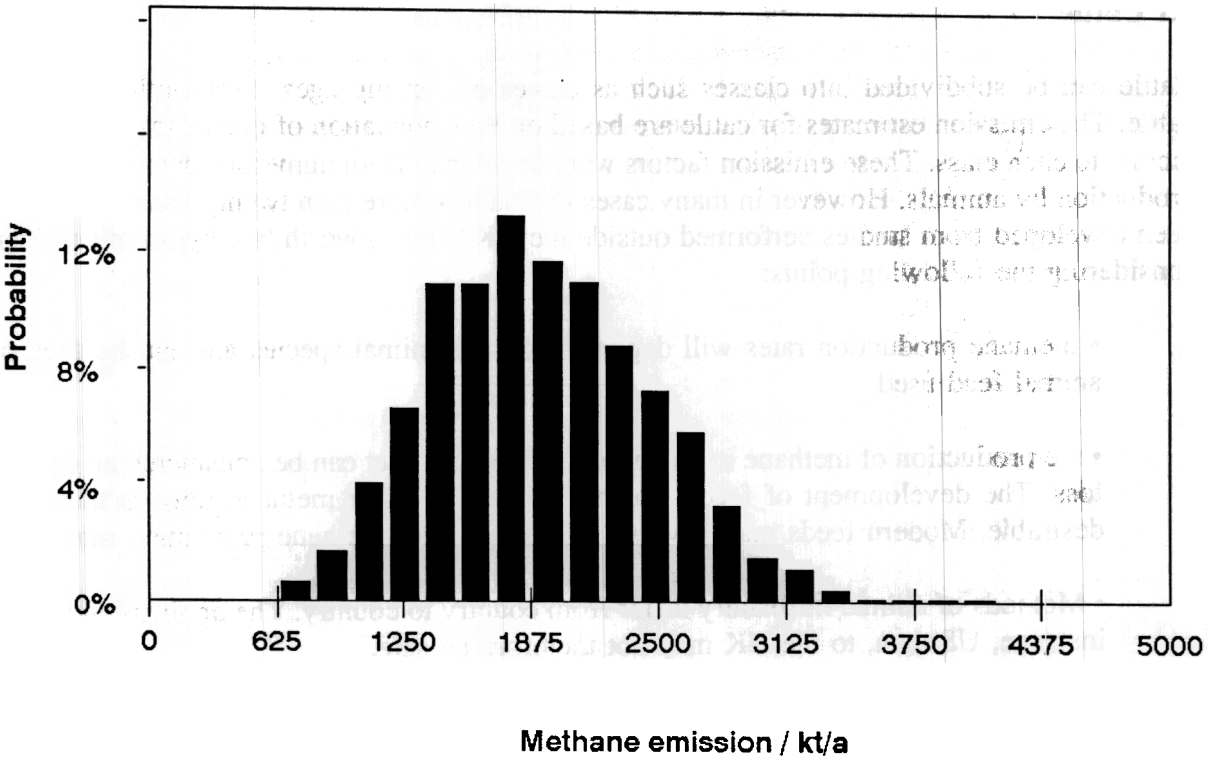


Figure 1. Uncertainty in emissions of methane from landfills

4 ANIMAL RESPIRATION

The estimates presented in this report are based on those produced by the Watt Committee, and are calculated using published methane emission factors (Crutzen 1986, Eggleston 1989), multiplied by data on numbers of animals published each year by MAFF. A +/-20% level of uncertainty has been estimated by the Watt Committee but this has not been tested and a higher number cannot be ruled out. The source paper referenced by the Watt Committee suggests an uncertainty of 15%. The simulation assumes that the probability distribution for the emission factors are triangular. A range is also used for the numbers of each animal. The actual emission could lie outside the predicted range as the inputs used to derive the estimates are untested and subject to a great deal of uncertainty. This is discussed in more detail in the following sections.

4.1 Cattle

Cattle can be subdivided into classes such as calves of varying ages, beef cattle and dairy cattle. The emission estimates for cattle are based on the application of one of three emission factors to each class. These emission factors were developed from numerous data on methane production by animals. However in many cases this data is more than twenty years old or has been developed from studies performed outside the UK. This is worth bearing in mind when considering the following points:

- methane production rates will depend upon the animal species and on the type of animal feed used.

- the production of methane in an animal's digestive tract can be considered an energy loss. The development of feeds which give rise to lower methane rates is therefore desirable. Modern feeds may have significantly lower methane production rates.

- Methods of animal husbandry differ from country to country. The application of, for instance, US data, to the UK may not therefore be safe.

- Feeding practices may have changed since the publication of the original source documents. In particular, average body weights for cattle and feeding levels may have changed.

In view of the above points, the uncertainty in estimates of emissions from cattle respiration would seem likely to be considerably higher than the 15% originally proposed, and probably also higher than the 20% suggested by the Watt Committee. For this study however a triangular distribution will be used, taking the Watt Committee emission factor as its most likely value and using a +/- 20% uncertainty to derive the limits. A normal distribution is used for animal numbers. For this study it is estimated that the standard deviation will be equal to 5% of the mean (which is equivalent to a 95% confidence level that the true value is within +/- 10% of the mean). The data used in the risk analysis is shown in Table 6.

Table 6 Data used to calculate emissions from cattle

Class of cattle	Emission factor (kg/animal/a)	Number of animals (millions)
Dairy in milk	T(76,95,114)	N(2.44,0.12)
Dairy not in milk	T(76,95,114)	N(0.33,0.017)
Beef	T(52,65,78)	N(1.67,0.083)
Bulls	T(52,65,78)	N(0.08,0.004)
Heifers in first calf		
for beef	T(52,65,78)	N(0.53,0.027)
for dairy	T(76,95,114)	N(0.2,0.01)
Others (> 2 years)	T(52,65,78)	N(0.68,0.034)
Others (1 - 2 years)	T(41,51,61)	N(2.58,0.13)
Others (< 1 year)	T(41,51,61)	N(3.36,0.17)

The estimates of methane emissions from cattle respiration is between 687 and 900 kt/a with a best estimate of 787 kt/a. A data quality of D is given to the emission estimates. The uncertainty is therefore high. In order to improve these estimates, more data is needed. Furthermore, the present practice of assigning one of three emission factors to each class of cattle defined (9 classes in total) in MAFF population statistics may also be over simplistic. Typical methane production rates for each type of animal should be developed.

4.2 Sheep and other animals

Similar arguments apply to emission estimates for sheep and other animals. There appear to be fewer data for sheep, pigs and other animals than exist in the case of cattle, and the variation in measured methane production rates is larger. Data quality is therefore low and a rating of D is given for these sources. As for cattle,it is hard to estimate a range of emission factors with confidence, so triangular distributions are used for the emission factors assuming a maximum uncertainty of +/-20% and normal distributions are again used for animal numbers. The data used is shown in table 7.

Methane emissions from sheep are estimated to be between 198 and 318 kt/a, emissions from other animals are between 18 and 26 kt/a. Together with cattle the model estimates emissions to be between 959 and 1196 kt/a with a best estimate of 1066 kt/a. This distribution is shown in Figure 2.

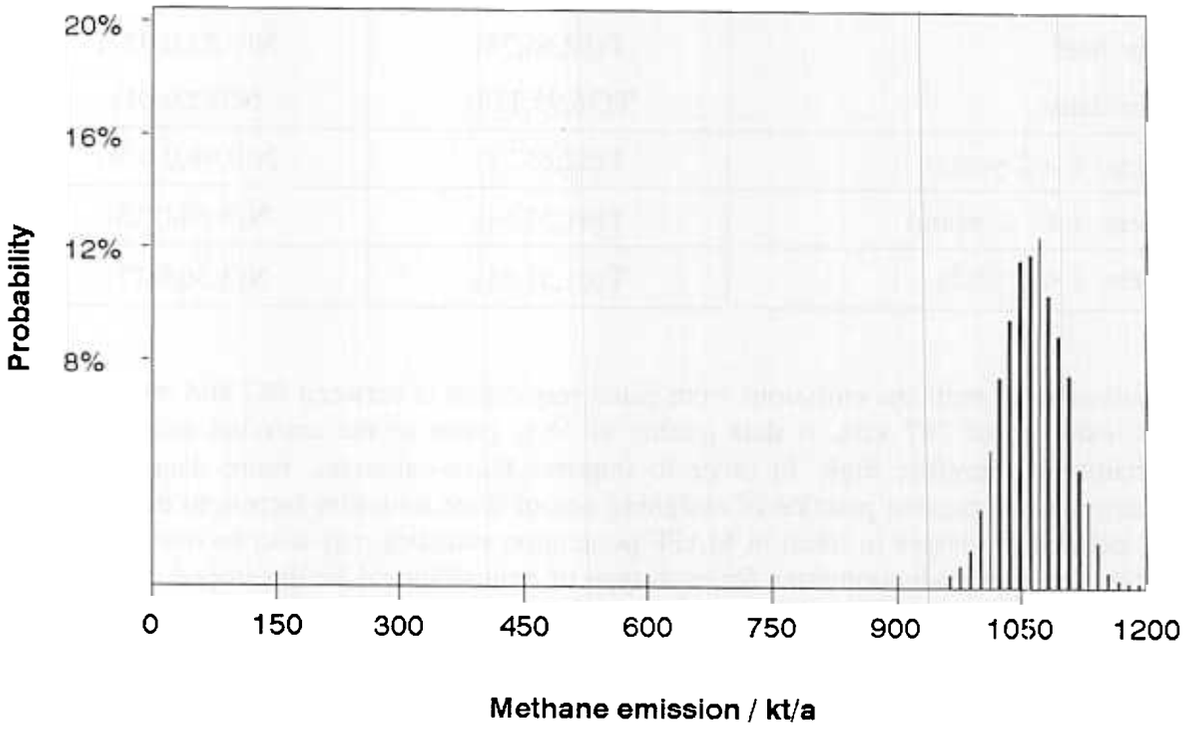


Figure 2. Uncertainty in emissions of methane from animal respiration

Table 7. Data used to calculate emissions from other animals.

Livestock	Emission factor (kg/animal/a)	Number of animals (millions)
Sheep		
- breeding	T(6.4,8,9.6)	N(20.4,1.02)
- others	T(6.4,8,9.6)	N(11.7,0.59)
Pigs	T(1.2,1.5,1.8)	N(7.60,0.38)
Poultry	T(0.04,0.05,0.06)	N(137.3,6.9)
Horses	T(14.4,18,21.6)	N(0.18,0.009)

5 ANIMAL WASTES

There is considerable uncertainty in the emission factors used to calculate emissions from this source. The maximum potential methane emission from animal wastes under ideal anaerobic conditions is considerably higher than estimates of actual methane production for typical treatment routes. The simulation uses the Watt Committee methodology but assigns uncertainty ranges to many of the inputs. Total waste arisings are calculated by multiplying numbers of animals by waste arisings per animal. Some of the waste from housed animals is stored as slurry, while the remaining manure from these animals, together with manure from free-range animals is not treated. Table 8 gives details of the ranges used in the waste arisings calculations.

The methane generated by this manure is then calculated using the equation below:

$$E = M_w \times VS \times B_o \times MCF \times C$$

where:

E = Emission of methane
M_w = Mass of waste
VS = Fraction of volatile solids in the waste
B_o = Potential methane generated by the volatile solids in m^3kg^{-1}
MCF = Fraction of theoretical methane generation which is actually emitted (methane conversion factor).
C = Mass of a cubic meter of methane at STP.

Table 9 shows the data used in the model. The values for volatile solids (VS) and the theoretical methane generation (B_o) are entered as single values. These values are given in a report by the OECD (1991) and were reported to be estimates based on measurement work.

These values have therefore been entered without ranges although their accuracy needs to be confirmed. The fraction of methane actually emitted (MCF) is subject to much greater uncertainty. The OECD report suggests that for slurried waste this will range between 10% for slurry stored for less than one month to 20% for slurry stored for more than one month. The Watt Committee suggests a typical value of 17%. The model used for this work will therefore use a triangular distribution centred on 17% with the range 10 to 20% (T(10,17,20)). Manure left in fields or stored in piles is estimated to have values for MCF of between 5% and 10% so a uniform distribution U(5,10) will be used. The OECD report does give a MCF of 90% for wastes stored in anaerobic lagoons (slurry stored in deep pits where anaerobic conditions will prevail). It is unclear whether this practice is actually used at all in the UK so it has not been taken account of in the model. However, it does show the potential for inaccuracy in the methane estimates if wrong assumptions are made about treatment of wastes.

Table 8. Waste arisings from livestock

Livestock	Numbers of animals (millions)	Waste per animal (t/animal/a)	Animals housed %	Waste stored as slurry %
Cattle				
Dairy in milk	N(2.44,0.12)	T(18.7,20.8,22.9)	50	80
Dairy not in milk	N(0.53,0.027)	T(11.2,12.4,13.6)	50	20
Beef	N(2.2,0.11)	T(11.2,12.4,13.6)	50	20
Others (> 2 years)	N(0.76,0.038)	T(8.9,9.9,10.8)	50	20
Others (< 2 years)	N(5.94,0.30)	T(4.9,5.5,6.0)	75	20
Pigs	N(7.60,0.38)	T(1.14,1.27,1.4)	100	75
Poultry	N(137.3,6.9)	T(0.025,0.028,0.031)	100	15
Sheep				
Breeding	N(20.4,1.02)	T(1.31,1.46,1.61)	0	0
Others	N(11.7,0.59)	T(0.72,0.80,0.88)	0	0
Horses	N(0.18,0.009)	T(5.91,6.57,7.13)	50	0

Table 9 Inputs for calculation of methane emitted from animal wastes

Livestock	VS (%)	B_o (m^3kg^{-1})	MCF for slurry %	MCF for manure not slurried %
Cattle				
Dairy in milk	12	0.24	T(10,17,20)	U(5,10)
Dairy not in milk	12	0.24	T(10,17,20)	U(5,10)
Beef	12	0.17	T(10,17,20)	U(5,10)
Others (> 2 years)	12	0.24	T(10,17,20)	U(5,10)
Others (< 2 years)	12	0.24	T(10,17,20)	U(5,10)
Pigs	10	0.45	T(10,17,20)	U(5,10)
Poultry	19	0.32	T(10,17,20)	U(5,10)
Sheep				
Breeding	23	0.19	T(10,17,20)	U(5,10)
Others	23	0.19	T(10,17,20)	U(5,10)
Horses	20	0.33	T(10,17,20)	U(5,10)

Emissions are estimated by the model to be between 329 and 598 kt/a with a best estimate of 466 kt/a. This range is shown in Figure 3. This uncertainty comes partly from uncertainties in waste arisings per animal but predominantly due to the uncertainty in the treatment routes used for those wastes and in the methane generated by those treatment routes.

Specifically, data is required to confirm the following:

- the waste production of each type of animal
- details of quantities of wastes treated in slurry systems or by other routes (including anaerobic lagoons, anaerobic digesters, spreading etc)
- details of storage times for slurries.
- testing of methane conversion factors for main waste treatment processes.

Due to the large number of uncertainties in the estimates of methane emissions from this source, a data quality of D is given.

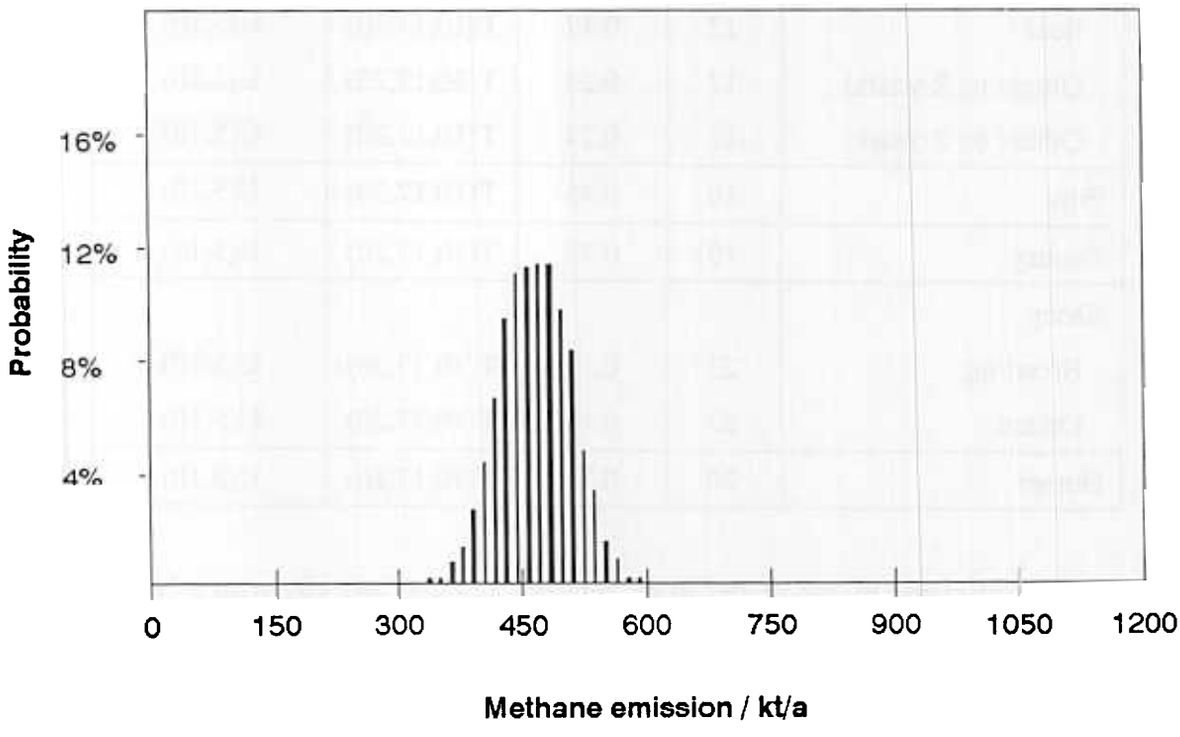


Figure 3. Uncertainty in emissions of methane from animal waste

6 COAL MINING AND DISTRIBUTION

The Watt Committee have produced emission estimates derived from measurements taken weekly at each colliery during the period January to June 1991. This data was converted to a mean emission per tonne of coal produced. The uncertainty in the emission factor is +/-15%, derived from the standard deviation in the measurements of deep mine emissions. A normal distribution is therefore used in the risk analysis with a standard deviation equal to 7.5% of the mean (ie 95% of values are within +/-15% of the mean). Coal production statistics are assumed to be accurate, therefore reported values are entered as single values. The inputs are summarised below:

Table 10 Data used to calculate emissions from mining

Emission source	Emissions from coal (Kg/t)	Mass of coal mined (Mt)
Deep coal mines	N(10.41,0.78)	73.36
Opencast mines	N(0.394,0.0296)	18.62

The calculation results in a best estimate of 771 kt/a with a range of 548 to 968 kt/a, as shown in Figure 4.

Further monitoring, carried out continuously at a number of mines should further improve the quality of emission estimates. Data quality for this estimate is therefore B at present but a data quality of A could be given if measurements were made continuously at each mine. Emissions of methane from open cast mines are estimated to be lower and are more closely related to the methane content of the coals being mined. Emission factors are based on measurement of a number of open cast coals. A data quality of B is given.

Mined coal will contain some residual methane, estimated by the Watt Committee to be 40% of the original content. This is equal to about 2 m^3/t. All of this methane is assumed to be lost during transport and storage of the coal. This could be tested by analysis of coals being fed to combustion plant. If in fact, methane is still present in these coals, then the methane emission from coal mining would need to be revised downwards slightly. It should be emphasized that this is a minor correction, as it refers to 40% of the methane contained within the mined coal and not 40% of the methane released by the mining of the coal. Most of the methane released comes not from the mined coal itself, but from the surrounding strata which are disturbed by the removal of the coal.

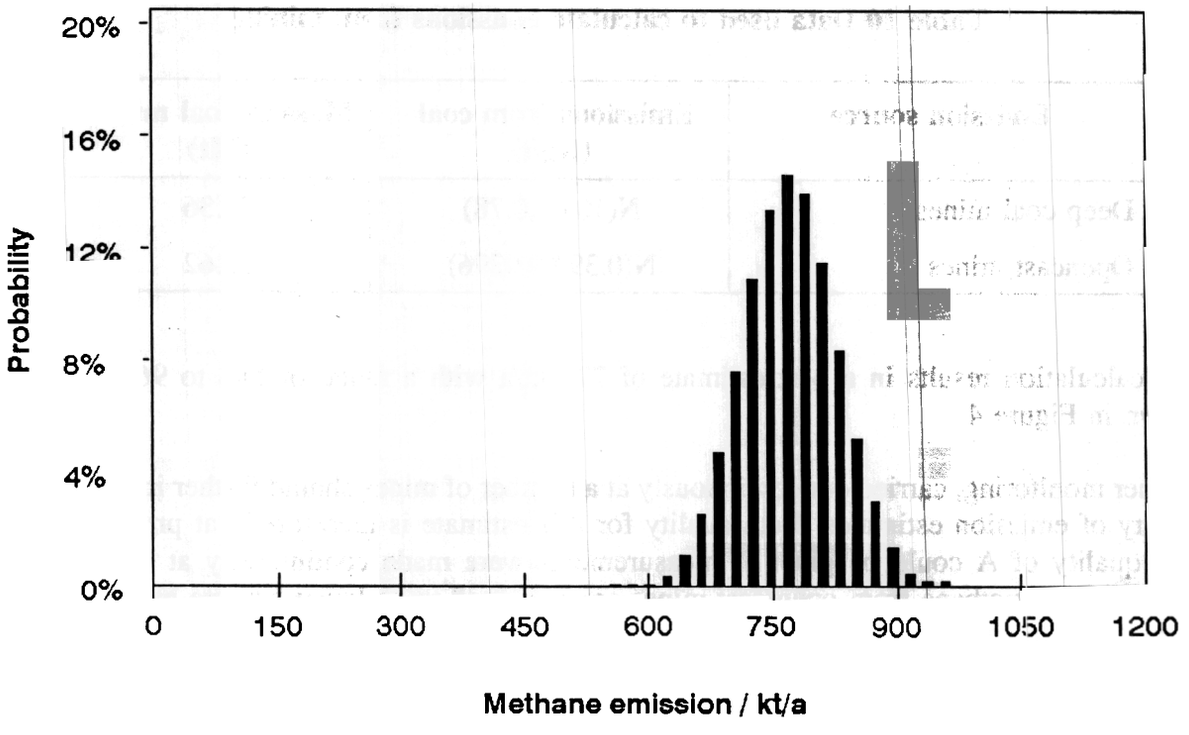


Figure 4. Uncertainty in emissions of methane from coal mining and distribution

7 GAS LEAKAGE

Emissions from the gas distribution network have been estimated by the Watt Committee. These sources of methane have been categorised as follows:

Transmission:
- Terminals
- Storage and LNG sites
- Transmission operations

Distribution:
- Low pressure (LP) mains and service leakage
- Medium pressure (MP) mains leakage
- Governers and holders
- Broken/purging mains etc

Commercial and industrial:
- Isolated releases
- Continuous long term release

Leakage past the meter:
- Commercial and industrial
- Domestic appliances
- Domestic carcassing leakage

Many of these sources are very small and the detailed methodology for the estimation of emissions given in the Watt Committee report is not repeated here. Emissions are all calculated as natural gas. Natural gas is 91.6% methane, therefore each emission would need to be multiplied by 0.916 to convert to methane.

7.1 Transmission losses

Losses of natural gas from terminals are estimated by the Watt Committee although no method is given. The figure also does not include fugitive losses. The losses from storage sites is based on estimates made by site personnel. Neither estimate is therefore based on measurements and so a data quality of D will be given.

Emissions from operations are predominantly from venting. The emission estimates for these sources are based on data from equipment manufacturers together with a small number of measurements from the venting of on line compressors. This estimate can be given a data quality of C; the remaining operations are given a data quality of D. The Watt Committee report that the level of uncertainty for this data is +/-30%. In view of the low data quality for these estimates, the model will use a uniform distribution for the estimates with +/-30% range around the Watt Committee estimates. The data is shown in Table 11.

Table 11 Estimates of methane emissions from transmission

Source	Estimated emission of gas (t/a)
Terminals	U(527,979)
Storage	U(226,420)
Operations	
Venting from control equipment and pipetraps	U(8941,16606)
Venting from compressors	U(810,1504)
Pipeline decommissioning	U(1492,2772)
Other	U(164,304)

7.2 Distribution

Multiple measurements of leakage from LP and MP mains in the UK have been made and these estimates can therefore be given a high data quality of B. This work has led to development of average loss rates per km of pipeline. These factors are multiplied by the total length of each type of mains in the distribution network.

The Watt Committee reports an uncertainty of +/-17% (at a 90% confidence level) for the low pressure mains, so this report will use a normal distribution with a standard deviation equal to 8.5% of the best estimate. The MP mains estimate is given a +/-20% uncertainty so a normal distribution with standard deviation equal to 10% of the best estimate will be used.

The remaining sources of gas leakage from the distribution network are governers, holders, purging of mains, broken mains and mains damaged by interference. Most of these sources emit very small amounts of gas and so the Watt Committee methodology for estimation of these losses will not be detailed here. Broken mains are the most significant source and emissions are calculated by assuming the measured leakage rate for 6" pipes occurs in all cases of broken mains and that leakage is detected and repaired within 8 hours. The average number of broken pipes over a six year period was used to derive an annual emission. This estimate may be exaggerating emissions from broken mains since most broken pipes are between 3" and 6". A low data quality rating of D is given to this estimate. The Watt Committee suggests a +/-75% range but in view of the possibility of overestimation for this source, a uniform distribution with ranges +50% and - 75% is used.

The remaining estimates are based partly on limited measurement work and partly on assumptions. A data quality of C is given to the estimates and the Watt Committee's suggested +/-50% range is used to derive a triangular distribution. Table 12 summarises the emission estimates for gas distribution.

Table 12 Emissions from gas distribution

Source	Estimated emission of gas (t/a)
LP mains	N(392100,33330)
MP mains	N(3990,399)
Broken mains	U(2120,12720)
Interference mains & services	T(269,537,806)
Purging mains & services	T(36,71,107)
Governers	T(355,710,1065)
Holders	T(939,1865,2798)

7.3 Commercial and industrial utilisation

The Watt Committee divides these into isolated, regular and continuous losses. The isolated emissions are very small (about 25 t/a) so the uncertainties in estimates of these sources will not be considered further. The regular emissions come from plant shut-downs and are calculated assuming one shut-down per hour, 10 hours a day, 5 days a week, 50 weeks a year. This is probably high for many types of boiler, one tenth of the number of shut-downs being a likely lower limit. This gives a total emission range of U(48,481).

Other regular emissions include valve proving U(2,92) and failed ignition which is given a range of U(43,128). All of these emission estimates are based entirely on untested assumptions and are therefore given a data quality of E.

Continuous losses include incomplete combustion (which will be dealt with as part of section 10, and leakage from pipes. Most installations are assumed by the Watt Committee to meet existing standards, with the remainder leaking at higher rates, giving a total emission of 5902 t/a. The Watt Committee assign a +/-50% uncertainty. This report will use a triangular distribution and the estimate will be given a data quality of D. Inputs to the risk analysis are summarised in Table 13.

Table 13 Emissions from industrial gas utilisation

Source	Estimated emission of gas (t/a)
Shutdowns	U(48,481)
Vent valve during proving	U(2,92)
Ignition failures	U(43,128)
Pipeline leakage	T(2951,5902,8853)

7.4 Domestic use

Losses are divided into losses from appliances such as cookers, fires and water heaters and losses from domestic pipework.

Based on measurements of four (not necessarily typical) central heating boilers, the Watt Committee report an average methane loss of 0.21 to 0.37% of gas consumption (with a midpoint of 0.29%). However these estimates are reported to produce excessively high results, hence the Watt Committee suggests a figure of 0.2%. This gives an emission estimate of 33 kt/a. For the present study, the original range of 0.21 - 0.37% has been assumed to give a good indication of uncertainty and a similar percentage uncertainty (+/- 27.5%) has been fitted to the best estimate above and a triangular distribution used. The estimate is given a data quality of C.

Measurements on two cookers were used as the basis of emission estimates for these appliances. Losses occur during ignition and extinction of the flame and there are also steady state losses during use. Usage patterns for cookers have been determined by a survey by British Gas so the estimates for this source have a reasonable level of confidence. A data quality of C is given and a triangular distribution with limits based on a arbitrary +/-50% uncertainty is used for the risk analysis.

Losses from gas fires are based on emission factors measured for cookers and assumptions on gas fire usage and gas consumption. The data quality is therefore lower (D), however a similar +/-50% uncertainty will be used to derive a triangular distribution.

Losses from water heaters are assumed to be similar to boilers, and similar assumptions on usage patterns are made as for boilers. The data quality is D and a +/- 50% uncertainty will again be used in the risk analysis.

The Watt Committee reports the results of a survey of gas leakage from pipes which suggests that leakage is zero in 56% of houses, up to $5m^3/a$ in 35% and above $5m^3/a$ in the remainder. The average leakage was $1.7m^3/a$ giving a total emission of 19969 t/a methane. This study will assign a data quality of C and arbitrarily use a +/-25% uncertainty in the risk analysis. Table 14 shows the emission ranges used in the model.

Table 14 Emissions from domestic use of gas

Source	Estimated emission of methane (t/a)
Central heating boilers	T(23943,33025,42107)
Cookers	T(1850,3700,5550)
Gas fires	T(1300,2600,3900)
Water heaters	T(300,600,900)
Pipework leakage	T(14977,19969,24961)

The overall total emission from the gas distribution network and gas leakage from users is between 338 and 564 kt/a with a best estimate of 454 kt/a. The range is shown in Figure 5. The uncertainty in this estimate is lower than for most of the other major sources of methane. Some considerable uncertainties remain for some of the smaller sources within the industry, such as transmission terminals and storage, broken mains and domestic appliances. The Watt Committee reports that some monitoring is underway so improvements to some estimates may be possible at a later date.

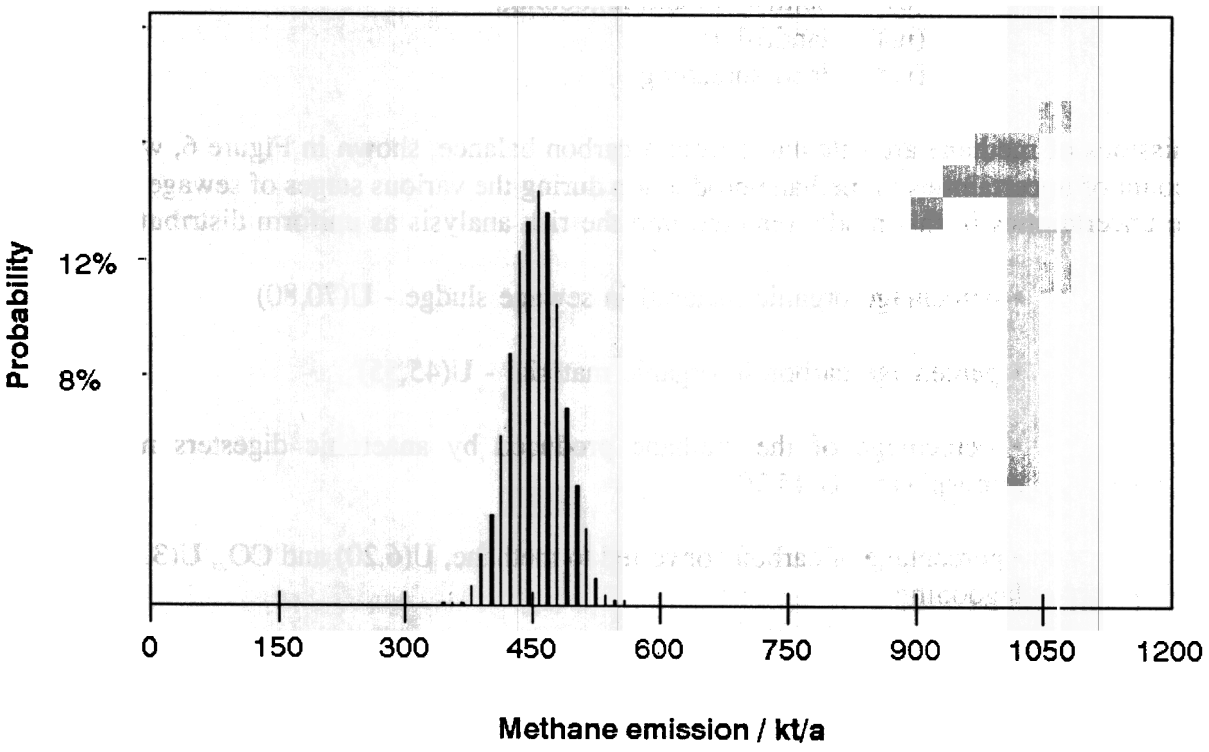


Figure 5. Uncertainty in emissions of methane from gas leakage

8 SEWAGE

Emission factors are taken from the work of the Watt Committee and are subject to large uncertainties. As shown in Figure 6, sewage sludge may be disposed of in one of four main ways - incineration, landfill, land spreading and dumping at sea (although the last will decrease due to new legislation). Alternative treatments, such as composting, are beginning to increase in importance. Methane can be emitted by sewage sludge disposed of by each route although the emission per tonne of sludge does differ considerably. The tonnages of sewage sludge disposed of by each route is therefore of considerable importance. UK arisings of sewage sludge and disposals have been estimated by the DoE (1992).

Emissions of methane from sewage sludge disposal arise largely from the treatment of the sludge before disposal. Only in the cases of landfill and land spreading is the disposal method thought to produce methane. Sources of emissions can therefore be summarised as:

(i) digestion,
(ii) settlement and lagooning,
(iii) landfilling,
(iv) land spreading.

Emissions of methane are calculated from a carbon balance, shown in Figure 6, which takes account of uncertainties in methane production during the various stages of sewage treatment. The uncertainties in this model (entered into the risk analysis as uniform distributions) are:

- percentage organic material in sewage sludge - **U**(70,80)

  percentage carbon in organic material - **U**(45,55)

- percentage of the methane produced by anaerobic digesters released to atmosphere - **U**(15,20)

- percentage of carbon converted to methane, **U**(6,20) and CO_2, U(3,10) during lagooning

- percentage of carbonaceous matter disposed of by each route:

Incineration	**U**(5,8)
Landfill	**U**(16,21)
Dumping at sea	**U**(30,35)

(The remainder is assumed to be spread or injected on land.)

- percentage of carbon converted to methane during the disposal of sewage sludge by land spreading **U**(5,10)

Emissions from landfilling of sewage are excluded from the totals for sewage because they are included in the landfill section.

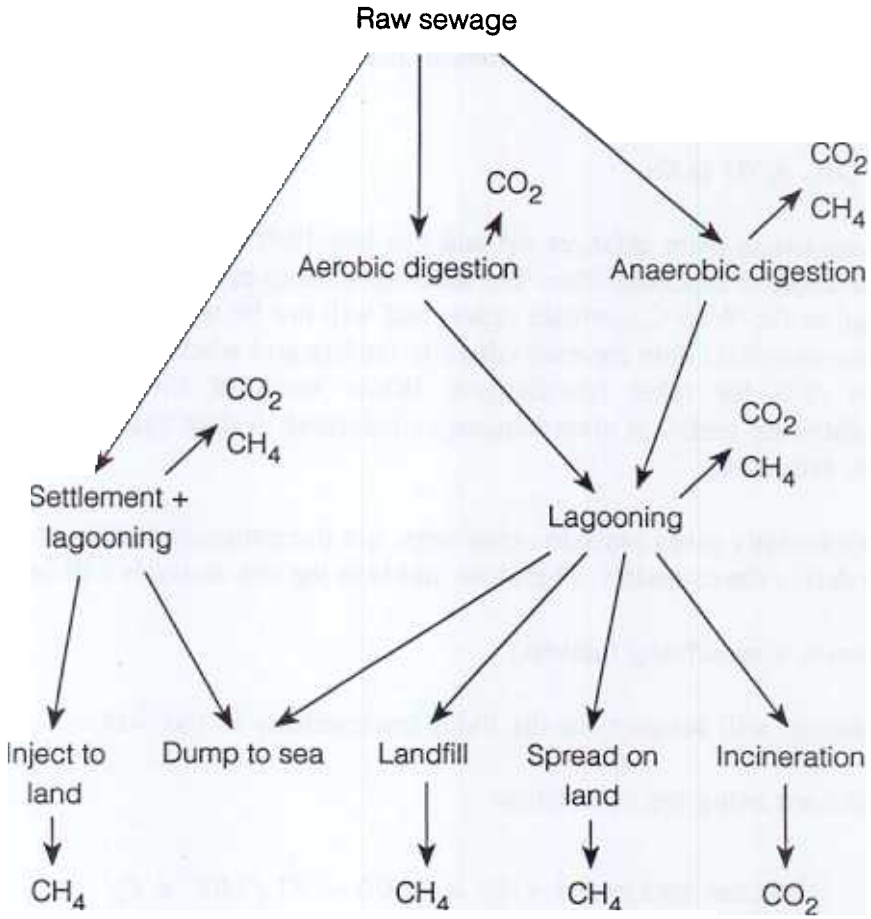


Figure 6. Treatment of sewage and associated emissions of carbon

Emission estimates are given a data quality rating of C in the Watt Committee report, although the emission factors do not appear to be based upon measurement work. We will therefore assign a data quality of D.

Emission estimates for sewage are between 45 and 107 kt/a with a best estimate of 71 kt/a (excluding landfilling of sewage sludge). Emissions from this source are therefore subject to a high level of uncertainty. Other recent estimates (Water Services Association 1993) fall within this range. To improve estimates better data is needed both for arisings of sewage and the fraction treated by different methods. There is also a need for measurements of the amount of methane generated from each different method of treatment.

9 OFFSHORE OIL AND GAS

Information on emissions from offshore oil and gas installations has been collected by the industry for input to the Watt Committee. The methods used to estimate these emissions were described in detail in the Watt Committee report and will not be reproduced here. The basic approach has been to collect data for each offshore facility and where this is not possible, to extrapolate from data for other installations. While some of the data are based on measurements, others are based on mass balance calculations. A data quality of C is therefore given to all these estimates.

The Watt Committee only gives emission estimates, not the emission factors and production statistics used to derive the estimates. Therefore inputs to the risk analysis will be in the form:

Best estimate x modifying factor(s)

The modifying factors will account for the likely uncertainties in the data.

Flaring was calculated using the calculation

$$Volume\ emitted = V_f \times (100 - F)/100 \times f_m$$

where

V_f = volume of gas flared
F = destruction efficiency of flares (%)
f_m = fraction of methane in the gas

Flare efficiency is reported by the Watt Committee to be usually in the range 98 - 99% with 98% used to calculate the best estimate. The risk analysis will assume that uncertainty in flare efficiency follows the distribution T(97.5,98,99) and that the volume of gas flared and the fraction of methane in the gas are known to +/-10%. The Watt Committee gives an estimate of 19.2 kt/a and hence the necessary modifications are

Emission = 19.2 x T(0.5,1,1.25) x T(0.9,1,1.1) x T(0.9,1,1.1)

Venting losses are calculated in a similar fashion to flaring.

$$Volume\ emitted = V_v \times f_m$$

V_v = volume of gas vented

An uncertainty of +/-10% has been assumed for the volume of gas vented and for the methane content of the gas. The Watt Committee's best estimate is 63.6 kt/a so the risk analysis derives estimates using the following equation:

Emission = 63.6 x **T**(0.9,1,1.1) x **T**(0.9,1,1.1)

Fugitive losses are calculated by the Watt Committee by summing the loss from each type of component:

$$Emission = \sum (N \times A \times f_m$$

N = Number of each component type
A = Emission factor for that component
f_m = Fraction of methane in the product stream

Component count studies have been carried out for a number of installations and this data is extrapolated to cover the remaining installations. Emission factors have been measured in a number of studies of these sources. Arbitrary uncertainties of +/- 5% and +/-10% are applied to the component counts and emission factors respectively. The Watt Committee best estimate is 11.2 kt/a and the risk analysis uses the following corrections

Emission = 1.2 x **T**(0.95,1,1.05) x **T**(0.9,1,1.1) x **T**(0.9,1,1.1)

The remaining sources are small in comparison and so an arbitrary +/-25% uncertainty is applied to these. The inputs to the risk analysis for all of these sources are given in Table 15. (The sub-sector best estimates do not add to the sector total because of a small rounding error).

Total emissions of methane from this sector are estimated at between 82 and 113 kt/a with a best estimate of 97 kt/a. These estimates are in broad agreement with values derived independantly by combining estimates of VOC emissions with the known methane content of gases from oil production.

Table 15 Methane emissions from minor sources in the offshore industry

Source	Estimate (kt/a)
Turbines	T(0.8,1,1.4)
Other combustion	0.1
Tanker loading Offshore Onshore	 T(0.5,0.7,0.9) T(2.0,2.7,3.4)
Storage Tanks	0.2

10 STATIONARY COMBUSTION

Emission factors for the domestic combustion of coal and solid smokeless fuel (SSF) are derived from a series of measurements on different combinations of fuels and heating appliances (Keeling 1992) shown below. A data quality of B is therefore given to these estimates. In the case of bituminous coal the mean emission factor is taken as the central value in a triangular distribution together with the minimum and maximum measured emission factors.

Bituminous coal / open fire (10 tests)

Minimum	1.6 kg/t
Mean	7.7 kg/t
Maximum	13.9 kg/t

Therefore the distribution is T(1.6,7.7,13.9).

The situation for solid smokeless fuels is more complicated because of the large number of fuel types and appliances. Emissions of methane from a variety of fuels and appliances have been measured (Keeling 1992). A mean emission factor, together with upper and lower limits have been derived from this data to give a triangular distribution T(3.4,6.8,12.5).

Fewer data exist for domestic combustion of other non-solid fuels. The table below summarises emission factors for methane taken from various sources (Watt Committee 1993, Veldt 1991, Passant 1993).

Table 16 Emission factors for domestic use of non-solid fuels

Fuel type	Emission factors (g/GJ)
Oil	3 7
LPG	1 10.6
Natural gas	3 4.3 15

The basis of these emission factors is uncertain in many cases so a data quality of D is given. For the risk analysis, uniform distributions will be used as follows:

Oil	U(3,7)
LPG	U(1,10)
Natural gas	U(3,15)

Some methane emission factors exist for industrial combustion taken from the same sources as for domestic combustion. These are shown in Table 17.

Table 17 Emission factors for industrial fuel use

Fuel	Source	Boiler type	Emission factor (g/GJ)
Coal	Electricity Supply	"Utility" > 50 MW	0.6 1
	Industry	"Industrial" < 50 MW	2.4 10
Oil	Industry	 Small plant Large plant	1 1.4 2.2 3
Natural gas	Electricity Supply	Large plant > 50 MW	0.8 1
	Industry	"Industrial" Small plant < 50 MW	1.4 1.5 4

The data quality is C and the following uniform ranges are used in the calculations:

Industrial

Coal	U(2.4,10)
Oil	U(1,3)
Gas	U(1.4,4)

Electricity supply industry (ESI)

Coal	U(0.6,1)
Gas	U(0.8,1)

The overall emissions from combustion sources are between 35 and 114 kt/a with a best estimate of 72 kt/a. The main areas of uncertainty are emissions from domestic combustion of solid smokeless fuels and natural gas.

11 REFINERIES

Estimates of emissions from refineries are based on new information from the United Kingdom Petroleum Industry Association (UK PIA). This estimate is based on the assumption that emissions of hydrocarbons from refineries will contain some methane which can be estimated by combining estimates of total hydrocarbon emissions provided by UK PIA with species profiles published by the United States Environmental Protection Agency (US EPA 1988). The risk analysis will apply an arbitrary range of +/- 25% to both the total hydrocarbon emissions and to the proportion of methane in the emission as shown in Table 18.

Table 18. Inputs for calculation of methane emissions from refineries

Source	THC emission / kt	Methane content
Flares	1	U(0.15,0.25)
Fugitive losses	U(14.25,23.75)	U(0.09,0.15)
Waste water treatment	U(15,25)	U(0.02,0.04)
Cooling water	U(2.25,3.75)	U(0.02,0.04)
Catalytic cracker	U(3.75,6.25)	U(0.19,0.31)

The best estimate for refinery emissions is 4 kt/a with a range of between 3 and 6 kt/a. The sector is therefore a minor source of emissions.

The US EPA gives data quality ratings of C or D to the species profiles. Hydrocarbon emission estimates used in the calculations also have data quality ratings of C or D. A data quality of D is therefore given to the methane estimate given here.

A typical refinery will have many potential sources of methane and a more detailed

description of sources within the refining industry is needed. Table 19 below gives a breakdown of sources of hydrocarbons within refineries. Emissions from these sources may contain methane. Other oil industry installations such as storage facilities and marine terminals could also emit methane and do not seem to be covered by the inventories at present available.

Table 19 Sources of hydrocarbon emissions at refineries.

Process losses Flares Fugitive losses from processes Process operations
Storage/tankage losses Storage of crude oil at refineries Storage of intermediates/products at refineries Storage of gases Storage of other materials Blending operations
Drainage and effluent treatment Waste water treatment Cooling water Interceptors
Distribution losses Loading/unloading operations Fugitive losses from distribution network

12 CHEMICAL INDUSTRY

Methane is important in the manufacture of ammonia, carbon disulphide and methanol. It may also be emitted from other processes as a product or by-product. Emissions from the chemical industry are based on an estimate of methane emissions from one major UK chemical manufacturer. This has been scaled up for the whole UK industry based on the production capacity for major bulk chemicals to give an emission estimate of 28 kt/a. Due to the fact that the basis of the original estimate is unknown and the uncertainty involved in scaling up in this manner, the estimate is given a data quality rating of D. No attempt has been made at this stage to quantify the uncertainty. A better understanding of methane sources within the chemical industry is necessary before emission estimates can be improved.

13 STRAW BURNING

Estimates of emissions from straw burning are derived using the Watt Committee method and data. The risk analysis uses uncertainties in the dry matter content of the straw, U(0.78,0.88),

and the proportion of the burnt carbon evolved as methane, U(0.007,0.013). A data quality of D is given to the estimate. The model estimates emissions to be between 12 and 24 kt/a with a best estimate of 17 kt/a.

14 TRANSPORT

Motor vehicle emission estimates are taken from the Watt Committee report. Vehicle emissions are estimated at 20 kt/a and no attempt has been made as yet to quantify the uncertainty for this estimate. Emission estimates are based on measurements of emission rates and species present. The NAEI estimates emissions of methane from other transport sources (shipping, railways and aircraft) to be 0.327 kt/a (in 1990). This is a very small source and has not been considered further in this work.

15 UNQUANTIFIED SOURCES

Other potential sources include secondary fuel manufacture, the use of bitumen, and abatement devices such as incinerators and biofilters. Natural sources such as mudflats, vegetation, marine and inland waters and soils also need emission estimates. Purely natural sources of methane are strictly outside the remit of the present work, but they cannnot be considered insignificant. The Watt Committee (1993) produced an estimate of 100 kt/a for the emissions from natural wetlands in the UK from various ground based measurements, although aircraft measurements suggested much higher figures of 380 to 1530 kt/a. The reason for this discrepancy is not understood.

16 RESULTS

Results of the risk analysis for total UK emissions are shown in Table 20. Those values that have not been quantified in this work are labelled "n/q". Values that are less than 1 kt/a are shown as "0". The best estimate for each source is given as well as the maximum and minimum value obtained. Data qualities are also given for each source. It will be noticed that in the case of minimum and maximum estimates, the subtotals and the total emissions are not equal to the sum of the individual emission estimates. This is because the subtotals and totals are calculated for each iteration and the minimums for individual sources are unlikely to occur on the same iteration. The probability distributions of the estimates for landfill, animal respiration, animal waste, coal mining and gas distribution are shown graphically in Figures 2 - 6. A combined distribution of emission estimates for all other minor quantified sources is shown in Figure 7 and the total emission is shown in Figure 8. The range of uncertainty is greatest for landfill and this has a major effect on the estimate of emissions from all sources. The range of uncertainty in the estimates generated for other sources is considerably smaller although it should be noted that in some cases, the data set used to generate the estimates was limited, and single values were entered for inputs where probability distributions would be preferable. Figure 9 illustrates the relative importance of the uncertainties by plotting the range of estimates against the best estimate for each sector. It demonstrates the dominance of the landfill uncertainty very clearly.

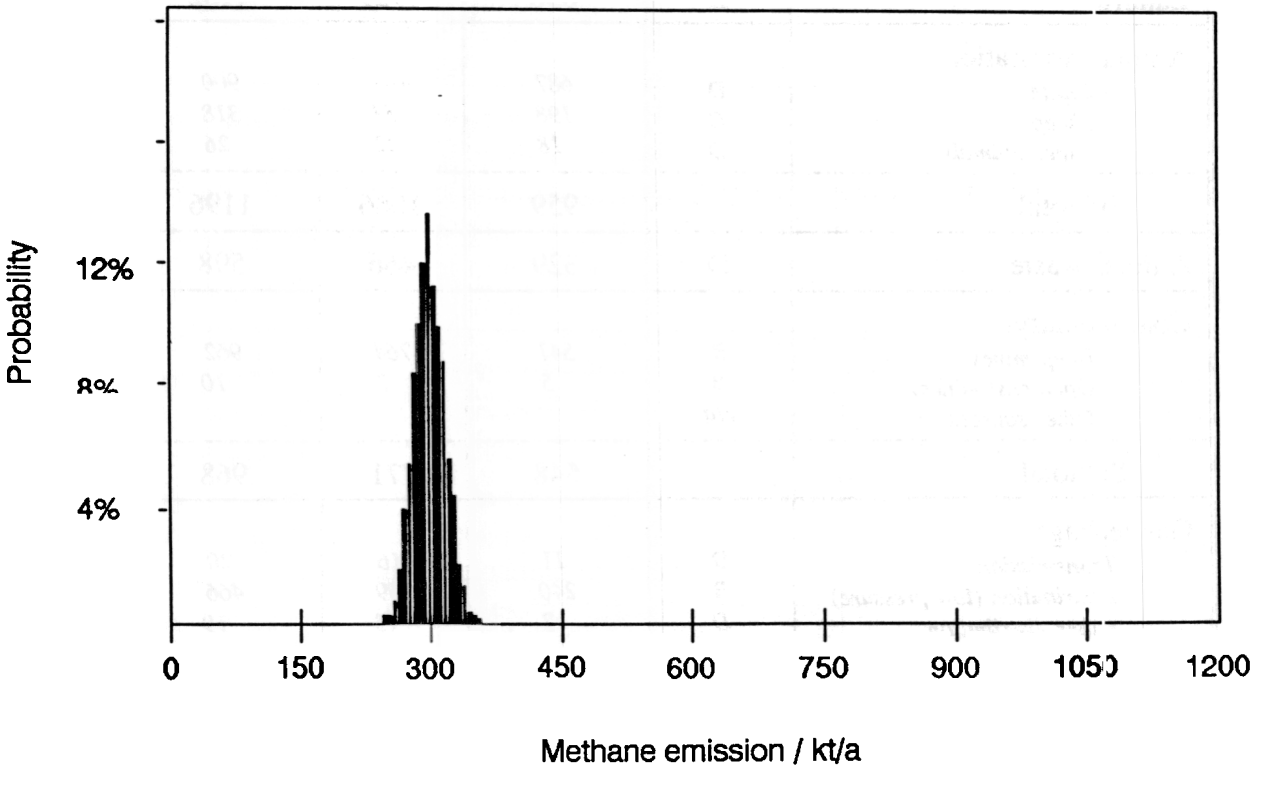


Figure 7. Uncertainty in emissions of methane from minor sources
This category includes all quantified sources apart from the major categories of landfill, animal respiration, animal waste, coal industry and gas leakage.

Table 20 Estimated emissions of methane from anthropogenic sources in the UK

Source	Data quality	Methane emission / kt/a		
		Minimum	Best	Maximum
Landfill	D	630	1929	4162
Animal respiration				
Cattle	*D*	*687*	*787*	*900*
Sheep	*D*	*198*	*257*	*318*
Other animals	*D*	*18*	*22*	*26*
Subtotal		959	1066	1196
Animal waste	D	329	466	598
Coal industry				
Deep mines	*B*	*541*	*764*	*962*
Open cast mines	*B*	*5*	*7*	*10*
Other sources	*n/q*			
Subtotal		548	771	968
Gas leakage				
Transmission	*D*	*11*	*16*	*20*
Distribution (low pressure)	*B*	*240*	*359*	*456*
Other distribution	*D*	*7*	*13*	*19*
Commercial and industrial	*D*	*3*	*6*	*9*
Domestic - heating	*C*	*24*	*33*	*42*
Domestic - pipes	*C*	*15*	*20*	*25*
Domestic - other sources	*D*	*4*	*7*	*9*
Subtotal		338	454	564

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Table 20 - continued

Source	Data quality	Methane emission / kt/a		
		Minimum	Best	Maximum
Sewage	D	45	71	107
Offshore oil and gas				
Flares	*C*	*9*	*18*	*27*
Venting	*C*	*53*	*64*	*75*
Fugitives	*C*	*9*	*11*	*13*
Other sources	*C*	*4*	*5*	*6*
Subtotal		82	97	113
Stationary combustion				
Domestic - coal	*B*	*7*	*31*	*55*
Domestic - SSF	*B*	*12*	*26*	*42*
Domestic - other fuels	*D*	*4*	*11*	*19*
Industry	*D*	*1*	*2*	*3*
Electricity supply industry	*C*	*1*	*2*	*3*
Other	*n/q*			
Subtotal		35	72	114
Refineries	C	3	4	6
Chemical industry	D	n/q	28	n/q
Straw burning	D	12	17	24
Transport	C	n/q	20	n/q
Other sources				
Food industry	*n/q*			
Fuel manufacture	*n/q*			
Other industry	*n/q*			
TOTAL		**3569**	**4995**	**7186**

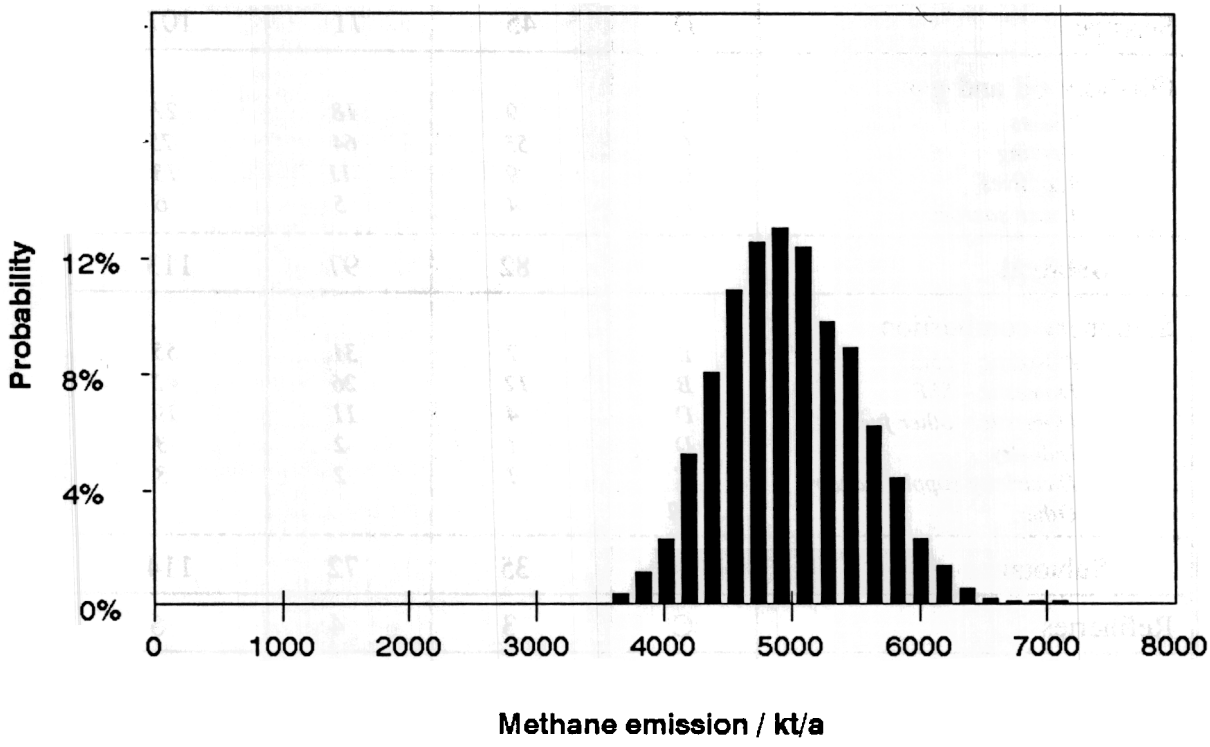


Figure 8. Uncertainty in emissions of methane from all sources

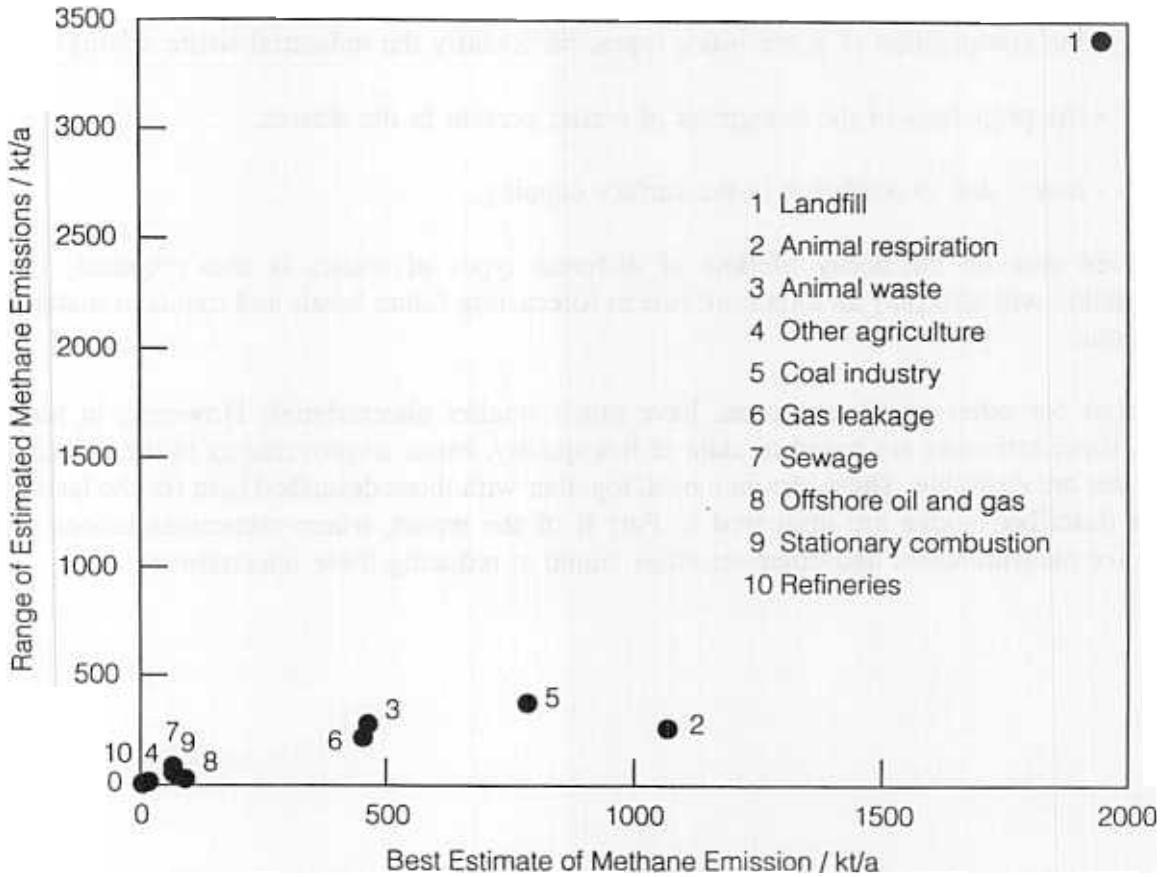


Figure 9. Sector uncertainty versus sector best estimate for all sources

17 CONCLUSIONS

Estimates of emissions of methane from anthropogenic sources are subject to considerable uncertainty. This uncertainty has been quantified using risk analysis and the quality of data used to derive estimates has also been assessed. The most significant contributor to the overall uncertainty is landfill. The best estimate of emissions from this source is 1929 kt/a, equivalent to 39% of the total. However, the risk analysis suggests that emissions could be anywhere between 630 and 4162 kt/a, equivalent to between 13% and 83% of the total (assuming that the best estimates for other sources are accurate). Improvements in landfill emission estimates are particularly reliant on better data concerning:

- the composition of some waste types, particularly the industrial waste arisings;
- the properties of the categories of matter present in the wastes;
- losses due to oxidation in the surface capping.

Improved data on the decay lifetime of different types of wastes is also required. This information will also play an important role in forecasting future levels and trends in methane emissions.

Estimates for other methane sources have much smaller uncertainties. However, in some cases, these estimates are based on data of low quality, hence improvements in the emission estimates are desirable. These requirements, together with those described here for the landfill sector described above are discussed in Part II of the report, where recommendations are made for measurements and other activities aimed at reducing these uncertainties.

Part II Section 1 - Techniques for the Measurement of Atmospheric Methane Emissions

The aim of this section is to review the techniques that have been used for measuring atmospheric methane and to show how they can be used to measure methane fluxes directly, and hence to improve estimated emissions. The techniques discussed fall into five categories.

i) Point Monitors and Flux Enclosures
ii) Optical Integrated-Path Monitors
iii) Range-Resolved Optical Measurements
iv) Micro-Meteorological Methods
v) Ambient Measurements and Modelling

1.1 POINT MONITORS AND FLUX ENCLOSURES

The most straightforward methods available for measuring ambient methane are the various types of point monitor (Department of the Environment 1992). Flux measurements can be derived from ambient concentration measurements by the use of a flux enclosure. This works by collecting methane in an enclosure and measuring the rate of increase of the methane concentration inside the enclosure.

1.1.1 Automated measurement of methane at fixed monitoring sites

The most widely used method for measuring methane is the flame ionisation detector (FID). Since it is sensitive to all hydrocarbons, if it is to be used to detect methane, it must be used together with a method for separating out non-methane hydrocarbons (NMH). The simplest method of doing this is a chemical converter which removes NMH at the input to the detector. Such a system has a sensitivity of approximately 100 ppb.

A more accurate and sensitive method is to pre-separate the methane using a gas chromatograph. Methane can be conveniently measured automatically over long periods at a fixed monitoring site using a gas chromatograph, with a flame ionisation detector. No pre-concentration is required, a metered volume of a few cm^3 of air being injected by an automatic valve directly onto a packed column. This results in ample separation with a high signal to noise ratio. Straightforward integration of the methane peak using standard software allows measurements to be logged locally on commercial data loggers, or to be integrated via a telephone link. Measurements are repeated automatically with a period of about 5 minutes. Individual measurements would resolve changes of 20 to 50 ppb, allowing the required sensitivity range of 100 ppb to 1 ppm to be achieved.

Periodic automatic injection of a standard air mixture would establish the stability of the system and provide the calibration factor. The accuracy of measurement would be of the order of 1 %. The technology employed in these measurements is well established, and is

similar to that routinely used in various applications of gas chromatography.

It is also possible to detect methane with a non-dispersive infrared detector (NDIR) with a similar sensitivity to the FID. There are also various types of solid-state and electro-chemical detectors that are capable of measuring methane, but they do not have the sensitivity required for these applications.

1.1.2 Flux Enclosures

The flux enclosure or "flux box" method is a technique that can be used under field or laboratory conditions to determine the flux of a gas to, or from a surface. In its simplest form an enclosure of known dimensions is placed over a surface and the rate of change of the concentration of the gas within the enclosure is measured with a point monitor.

The simplest type of box enclosure is a closed system in which emitted gases are simply allowed to accumulate. A more sophisticated design uses forced ventilation to flush the emitted gas through the enclosure. The flux is then calculated from measurements of the concentration of the target gas at the inlet and the outlet of the enclosure.

Hutchinson and Mosier (1981) have discussed the advantages and disadvantages of these two systems for studies of N_2O emissions from soils. The principal advantage of a forced ventilation system is that gas concentrations resembling those in the undisturbed environment are maintained. If there is a diurnal variation, or temperature dependent emission of the gas, the use of this type of system may be particularly advantageous. The major disadvantage is that instruments of high sensitivity are necessary as the difference between the inlet and exhaust concentrations may be very small. Careful calibration of the two instruments is also necessary (unless a single instrument with a line switching system is used).

Various measurements of fluxes of methane have been made using the box method. The following examples have all been taken using closed systems. Seiler *et al.* (1984) used a box chamber to measure the flux of methane from individual termite nests. (Termites are an important component of the global methane cycle (IPCC 1990).) An aluminium frame was covered by a plastic bag and air was removed via a septum into a syringe for analysis by gas chromatography. Whalen and Reeburgh (1990) used flux boxes on Arctic tundra in Alaska and the gas collected from a syringe sample was analyzed by gas chromatography. Morrissey and Livingstone (1992) also made measurements using the box method in the Alaskan Arctic tundra. The minimum average detectable emission rate given by Morrissey and Livingstone for methane was 3.3 $mg/m^2/day$ and 0.2 $mg/m^2/day$ by Whalen and Reeburgh (1990).

In order to achieve accurate results, it is important to establish whether the presence of the enclosure alters the emission rate at the surface. For example, the temperature within the enclosure may increase with respect to the ambient atmosphere, which may change rates of deposition or evolution of the gas. This is particularly important if surface microbiological conditions are important. For reactive or soluble gases, it is also important that significant deposition of gas to the enclosure surface does not occur. Finally, it is important not to compress or disturb the surface, as this may alter the anaerobic generation of methane.

The flux box is widely used for landfill emission measurements made according to US EPA-

approved methodologies (Eklund 1992) with a sensitivity of 1 $\mu g\ m^{-2}\ min^{-1}$. Typical accuracies obtainable are of the order of a few percent, depending on the details of the system used. In order to overcome the problems of spatial homogeneity with landfill methods, it is recommended that the average from six boxes is used to form the estimate for any specified area.

1.2 OPTICAL INTEGRATED-PATH MONITORS

There are a number of commercially-manufactured instruments available that are capable of measuring integrated-gas concentrations over paths of up to 500 m. These include Fourier Transform Infrared (FTIR) spectrometers, tunable-diode laser spectrometers and oscillating-filter spectrometers. The first of these (the FTIR spectrometer) has been used for direct emission flux measurements (Kirchgessner 1991), but is expensive and can be subject to interference from other atmospheric gases. The other techniques have been used widely. In particular, they have been used for a number of measurements of direct relevance to inventory verification (Milton 1992 and Partridge 1986).

The general principle of optical integrated-path monitors is that a beam of light is transmitted from the monitor into the atmosphere by a telescope and is then returned by a retro-reflecting mirror. The returned light is analyzed in order to determine the extent of any absorption caused by gases of interest. The instrument is then calibrated with a cell containing a known concentration of the target gas. Simultaneous measurements of the wind speed enable an integrated-path measurement to be integrated up to give a value for the average flux of gas across the light path. If a system of masts and mirrors is used to make these measurements at various different heights, the total flux across a vertical plane downwind of a source can be measured, from which the emission rate can be calculated.

An optical integrated-path monitor using the oscillating filter technique has been used to measure methane emissions from cattle (Milton 1992). Measurements were made of the speed of the wind and the excess methane concentration at several heights downwind of a field of bullocks. Accordingly the total flux of methane from the field was estimated, giving a figure for the total methane emitted by the cattle. This is an important measurement for the inventory as the specific emission of cattle is a quantity that is otherwise difficult to measure without disturbing the animals, and is a significant source of uncertainty in the inventory.

The application of these techniques to the measurement of methane fluxes from landfill sites has also been demonstrated. In two separate measurement demonstrations (Partridge 1986), a tunable diode laser spectrometer was used to make integrated-path measurements along a series of light paths just above ground level in various different directions across a site. In this way the heterogeneity of emissions across the site was mapped. Measured concentrations varied between 1 and 40 parts per million according to the wind conditions. In addition, measurements were made of the total flux of gas from the site by using a light path downwind of the site and at various different heights using mirrors supported on masts. These measurements were combined with wind speed measurements to give the total emission, a quantity which is difficult to obtain by any other direct method. The major sources of inaccuracies in this approach are the errors in the windspeed measurement and the flux of methane outside the area defined by the masts. Sensitivities of 0.1 parts per million have been

achieved over a 75 m path. The total error was estimated to be not more than about 15%.

For the purposes of improving the UK methane emissions estimate, this technique is most applicable to landfill. It has the advantage of being less expensive than the DIAL technology described in the following section, while it should provide flux data of comparable quality, as long as it is possible to set up suitable masts. The lower cost of the equipment makes deployment over extended periods feasible, which could allow monitoring of emissions under a wide range of meteorological conditions.

1.3 RANGE-RESOLVED OPTICAL MEASUREMENTS (DIAL)

It is possible to perform range-resolved measurement of methane in the atmosphere using the Differential Absorption Lidar (DIAL) technique. This technique has the advantage that it is able to make measurements without the use of a retro-reflecting mirror and can therefore make three-dimensional measurements of the space surrounding a source.

The principle of the system is that a short but high-powered pulse of radiation is fired into the atmosphere along a particular path while measurements are made of the light that is scattered back to a telescope mounted adjacent to the source. The back-scattering is caused by particulates and aerosols in the atmosphere, and the precise point at which the light is scattered is determined by measuring the time taken for the light to return to the receiver.

Two different wavelengths are used, one of which is absorbed by the gas of interest and the other of which is not, so that the concentration of the gas at a particular point along the light path is determined by comparing the amounts of light back scattered at the two different wavelengths. This allows range-resolved measurements of concentration to be made at distances of up to 3 km with a resolution of 10 m. Figure 10 is a schematic diagram of the method used to measure the cross-section of an emission plume in two dimensions by making measurements over several different lines of sight in rapid succession. The total flux of the gas is determined by monitoring the wind speed perpendicular to the cross-section chosen. An emission measurement for a complete site can be made by repeating the measurement up and down wind of the site.

The technique can be applied in the ultraviolet, visible or infrared spectral regions to investigate different species. Gaseous hydrocarbons such as methane are most easily measured in the infra-red using wavelengths around 3.3 μm. A typical application of the DIAL technique is the measurement of methane emissions from an onshore gas-processing terminal, described by Woods (1992). In this case measurements were made not only of the total flux from the site, but by careful choice of the light paths, the fluxes from separate components within the site were estimated. By using the system at different wavelengths it was also possible to perform a check for unexpected emissions of other hydrocarbon species.

DIAL is a very sensitive technique; it is possible to detect concentrations as low as a few parts in 10^8. The accuracy of the flux measurements is not limited by the DIAL component of the measurement but by the measurements of the wind speed. The resulting flux figures have uncertainties of typically 15%.

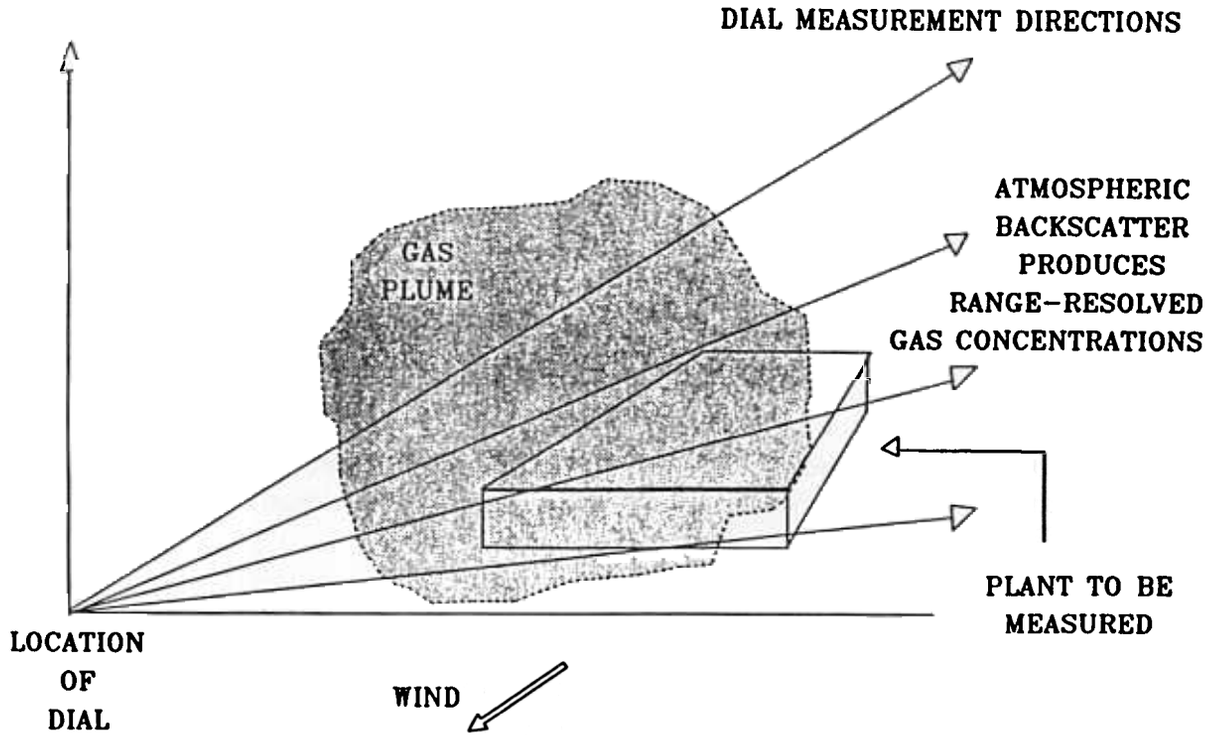


Figure 10. Methodology used for measuring fluxes by Differential Absorption Lidar (DIAL)

The major applications of DIAL have been to large industrial plant in the petrochemical and energy industries, but similarly it could be used for any reasonably compact source of methane (up to a spatial scale of typically a kilometre) where a complete picture of an emission plume is needed. Its drawback is the high capital cost of the equipment and the highly trained staff required for successful operation.

1.4 MICRO-METEOROLOGICAL METHODS

As the wind blows over an extensive area of uniform surface characteristics, an internal boundary layer develops in which the structure of the wind and turbulence, and vertical fluxes of momentum, heat, water vapour and trace substances are well adjusted to conditions at the surface. Within this layer, the vertical fluxes are constant with height and equal the fluxes at the surface, so that measurement of the vertical flux of methane at any height within the internal boundary layer provides information on the flux at the surface. Two general micrometeorological methods have been applied for the measurement of fluxes of trace substances at uniform surfaces; the gradient method and the eddy correlation method.

The gradient method requires measurement of the difference of concentration with height, and also the variation of wind speed, air temperature and, optionally, water vapour concentration. The method is well established, having been used to measure fluxes of carbon dioxide, sulphur dioxide (Garland 1977), ozone (Leuning *et al.* 1979) and particles (Garland and Cox 1972).

In essence, the method depends on the multiplication of the concentration gradient (dc/dz) by an eddy diffusivity (K) to obtain the flux (F) in a manner analogous to Fick's law for molecular diffusion in gases:

$$F = - K (dc/dz)$$

In practice, K is obtained from measurement of the profiles of wind speed and temperature with height, by reference to empirical relationships established in careful series of field measurements (e.g. Dyer 1970; Webb 1970; see also Garland 1977).

The concentration at each height is subject to short-term fluctuations, due to eddies of enriched air rising from close to the surface, and relatively depleted air from above, and in consequence the gradient at each height must be averaged over a time period long compared with the frequency of the dominant eddies; in practice it is usually convenient to average measurements over periods of 10 to 60 minutes. Interpretation of the measurements becomes difficult if the averaging time is too long, since the atmospheric conditions may change significantly.

Businger and Delaney (1990) provide guidance on the resolution required to detect a given upward flux of methane. The mean methane flux over the UK is expected to be of order 4 Mt/a from an area of order 10^5 km^2, hence the mean rate is of the order 10^{-6} g m^{-2}s^{-1}. Such a flux would give rise to concentration differences of order 1 ppb of methane. In practice one would normally be concerned with fluxes (and therefore concentration differences) of at least

an order of magnitude larger.

To apply the gradient method, measurements would be made close to the surface of interest. The well-adjusted internal boundary layer is expected to have a depth of about 0.01 of the upwind fetch (i.e. the distance that the wind has travelled over the uniform surface of interest before reaching the instruments) and measurements would be confined to that layer. Thus, if a uniform source area extended 1 km upwind, measurements would be confined within about 10 m of the surface. Measurement at several heights would be advisable to confirm that the wind, temperature and concentration profiles conform to the expected pattern, and give confidence in the application of the method. The range of heights used to derive the concentration gradient would be made as wide as practicable, noting that the lowest measurement level should ideally be several times the height of the roughness elements above the surface, and the uppermost level should be well within the internal boundary layer.

Davies and Dollard (1992) have shown that the gradient method is practicable for methane measurements, and obtained results generally consistent with the box method.

In contrast, eddy correlation methods require measurements at only one height, but care must be taken that the location is well within the internal boundary layer. The flux is obtained by calculating the correlation between concentration and the vertical component of wind velocity. The method is absolute, requiring no empirical relationships, but does require assurance that the instruments are fast enough to respond to the eddies which transport methane from the surface. For accessible heights within the internal boundary layer (1 to 10 m) this is likely to require instrument response times of order 0.1 to 1 s. The method has been applied for a number of tracers, including heat, sulphur dioxide (Galbally and Garland 1979), carbon dioxide and ozone.

Fan (1992) applied the method to CH_4 and CO_2 over sub-Arctic tundra, using a laser-based infra-red absorption method to measure methane concentration with a fast response time at a height of 1.2 m. They also measured the methane profile so that they could correct for storage of methane within the air column below 12 m. A fast flame ionisation detector was used to measure total hydrocarbon fluxes. Methane accounted for nearly all the concentration and flux.

In principle, the eddy correlation method is not influenced by instrument noise, provided it does not correlate with the short term variations from the mean of the vertical component of the wind velocity. The methane sensor is required to resolve concentration changes of order 1 ppb to measure fluxes of the order of the mean UK flux, and instrument noise and variations in upwind sources may introduce variations which may or may not correlate in short measurement runs.

The eddy correlation method can also be applied to measure fluxes over large areas, when mounted on an aircraft. The aircraft is flown over level flight paths. The output of the method is an estimate of the mean flux over an area sampled by the flight path flown.

1.5 AMBIENT MEASUREMENTS AND MODELLING

This technique uses ambient measurements made with one of the instruments discussed in section 1.1, together with a modelling technique to relate the measurements back to the emission rate at the source.

For "point" sources such as industrial plant or sewage works a simple approach is to measure the concentration at ground level downwind of the source, at sufficient distance to assume that any plume from a lofted release point has diffused to ground level. The emission rate can then be estimated by inverting the dispersion problem, using, for example, the Gaussian model (Pasquill and Smith 1983).

Application of the model requires adoption of empirical values for the dispersion parameters that describe the width and depth of the plume as a function of distance downwind. It also requires that the effective height of release (making allowance for any buoyancy of the plume) can be estimated so that measurements can be confidently made within the plume and that the wind speed at an appropriate height can be measured or estimated. The uncertainty in combining all of the empirical estimates may easily exceed an order of magnitude. The estimation of the long-term mean emission from sequential measurements of concentration at a small number of points in the vicinity of an isolated major point source may perhaps be achieved with better precision (a factor of two), by averaging over a number of measurement periods.

Similar approaches using ground-based point monitors can be applied to the plumes from large heterogeneous area sources such as cities. Measurements of non-methane hydrocarbons downwind of London from an aeroplane (Blake *et al.*, 1993) suggest that the methane plume from the capital city could also be measured in this way. Again, a combination of the airborne and ground based measurements might be used. The results of such a study could possibly provide verification for a part of the UK inventory.

In principle, this technique could be extended to the UK as a whole by making measurements at one or more fixed points. However, emissions from mainland Europe would make it difficult to separate the national emissions from those from the continent. Simmonds and Derwent (1991) used measurements at Mace Head (on the West Coast of Ireland) to estimate the emissions from Europe. They averaged data by wind direction into 45° sectors, and so were unable to distinguish the U.K. from the Low Countries and Germany.

A different, but related technique is to measure the ground level concentrations within a distributed source and to relate the results to the total source strength. Chamberlain *et al.* (1979) were able to relate the long-term mean concentration of lead in urban areas to the motor vehicle source. Continuous or periodic measurements at a small number of sites would be used to ensure that representative concentration measurements were obtained. It would also be necessary to make measurements of the background concentration outside the selected city.

Some preliminary modelling would be advantageous for either application as it would allow an estimate to be made of the influence of spatially heterogeneous emissions on the results. This would aid decisions on the number and location of measurement stations.

1.6 APPLICABILITY OF MEASUREMENT TECHNIQUES

The techniques discussed above fall into two groups. There are three techniques which basically rely on measurements on an emitted plume. The first two of these are the optical integrated-path monitors and the DIAL, which rely on "capturing" a cross-section of the plume with their light beams. The third "plume-based" technique combines measurement with point monitors in the plume with calculation by the inverted dispersion method. These three techniques are all applicable in principle to "point" sources such as landfill sites, industrial plant or sewage digesters. In practice, one of the optical techniques will give more accurate emissions estimates in situations where all of the plume can be captured.

For large, heterogeneous area sources, the point monitor and calculation techniques are more applicable. If natural sources and the background could be accounted for, it should be possible to obtain a value for the total contribution from an urban area and hence to verify a small section of the inventory. The drawback to this approach is the limited accuracy which will limit its applicability to inventory verification.

Box methods and micro-meteorological methods also have similarities. Box methods are most applicable to emitting surfaces such as landfill sites or wetlands, while the micro-meteorological methods require a large upwind area, which makes them ideal for wetlands, but of less use for typical active landfill sites, which generally do not have a sufficiently large uniform area.

In some situations, it is most likely that a combination of the above techniques will be the most cost-effective approach to the measurement problem. This is particularly so when emissions vary spatially or temporally, which is known to be a major issue in obtaining accurate estimates of total emissions from landfill sites. A technique such as an integrated-path monitor or DIAL could be used to obtain accurate total emission figures on a few days per year, while point monitors placed around the site could run continuously over a longer period. Provided there was reasonable confidence that the point monitors reflected the total flux, then a reliable estimate could be obtained for the total annual emission.

1.7 SUMMARY

The capabilities of the various techniques are summarised in Table 21.

Table 21 Direct Emission Measurement Techniques

Technique	Applications
Integrated-Path Optical Monitors	Measures cross-section of a plume from small sources e.g. landfill or industrial plant. Accurate to typically 20%. Able to produce spatially integrated measurements over extended sources.
Range-Resolved Optical Monitors	Measures cross-section of a plume. More versatile than integrated path, but more costly. Accuracy around 20%.
Flux Box	Measures emissions from surfaces. Applicable to landfill, but spatial heterogeneity is a problem. Has been used widely for measurements of landfill emissions. Accurate to a few percent.
Micrometeorological Techniques	Measures very low rates of emission from uniform surfaces. Most applicable to wetlands, as landfills are probably too uneven.
Ambient Measurements and Inversion Calculation	The only feasible technique where large area heterogeneous sources are to be measured. Unproven accuracy, possibly only a factor of two.

Part II Section 2 - The Application of Direct Measurements to the Improvement of the Estimate of UK Methane Emissions

An analysis of the uncertainties in the estimate of UK methane emissions from known sources was presented in Part I. In this chapter, we discuss how these estimates could be improved by the use of direct emission measurement techniques either on their own or in conjunction with modelling techniques. The discussion emphasises the landfill sector both because it is the most complicated problem and because it is the largest uncertainty in the estimate of total UK methane emissions.

2.1 LANDFILL

Methodologies for estimating gas emissions from UK landfill are poorly developed. Most approaches to date, including the one presented in the first part of this report, take the same basic approach. They make use of limited surveys of waste arisings and composition and estimates of the methane production potential of the material placed in landfill sites to estimate the total amount of methane generated. This figure is then corrected for the amount of methane that is oxidised or exploited for energy and the remainder is assumed to escape to the atmosphere. Generally, there has been little verification of these models by direct measurements of *emissions* from landfill sites. The measurements that have been carried out have concentrated on other issues, such as health and safety or gas exploitation. It is the aim of this section to examine how estimates of UK landfill emissions could be improved by the use of direct emission measurement techniques.

There are two possible approaches to making such improvements. They are not mutually exclusive, but have a considerable amount in common in the way they are based on a semi-empirical model of the methane emission process. The first starts with existing models and uses measurements to improve the estimates of critical parameters used in the models. The second also starts with existing models, but uses them as a basis for targeting direct emission measurements and for calculating the UK total from measurements performed on a selected set of sites.

2.1.1 Improvements to the Estimate of Landfill Emissions based on Modelling

The first approach builds on existing models, such as that presented in Part I. It uses a coordinated programme of laboratory experiments and ambient measurements to improve the accuracy of the parameters used. The detailed analysis of uncertainties in Part I shows which of the model parameters are most urgently in need of improvement. For example, there is a large uncertainty in the values used for the specific methane generation parameter. These have been determined by laboratory experiments and there is some doubt as to whether such values apply to the processes that take place in a real landfill. Another uncertainty is the degree of oxidation that occurs within the landfill cap. Both of these uncertainties could be effectively reduced by direct emission monitoring.

The major problem with improving parameter values by this approach is the difficulty, even

when measuring a well-characterised landfill site, of extracting a reliable value for one particular parameter. This is because the emission actually measured will also depend on many other parameters, each with their own range of uncertainty.

This modelling-based approach relies on accurate data for total waste arisings. In particular, it requires accurate data for waste arisings over a considerable period of time, since it is known that methane emissions continue for several decades. The models also need to use the quantities of different types of waste placed in landfills, as this is crucial for the calculation of the methane generation. Much of this data can only be obtained through extensive studies into the contents of all existing (including closed) landfills.

Finally, in addition to the benefits of using this approach to improve on individual parameter values, it would also be beneficial to verify the complete models by direct emission measurement.

2.1.2 Improvements to the Estimate of Landfill Emissions based on Measurement

It is not possible to make direct measurements of the emissions from all 12000 landfill sites in the UK. It is therefore necessary to adopt a methodology for identifying representative sites and extrapolating up from them to form a total for the whole UK.

The most efficient way to do this is by "stratified sampling". This is done by allocating the most important landfill sites to a limited number of categories or strata, according to the parameters that have the strongest influence on emissions. Possible stratifications could be based on the quantity and age of waste in place, the hydrogeology or design of the site. Measurements would then be performed on a set of sites in each category. In this way an emission factor is obtained which can be applied to all sites in the category. Scaling up from a few sites to the total for the whole category requires knowledge of the waste in place in all sites in the category.

Although there are databases in existence containing information about specific groups of landfill sites, there is no single source that covers all of the information required for estimating methane emissions from all UK landfills. The following sections give our proposal of how the uncertainty in emissions from UK landfills could be reduced by direct emissions measurement based on a stratified sampling approach using currently available data and taking account of other activities being carried out on waste disposal.

2.1.3 Stratifying UK Landfill Sites

The first step in establishing a stratification would be to obtain an estimate of the quantity of degradable waste in the UK. Referring to the data in section 3.1.1 of Part I, we calculate that annual UK *degradable* waste arisings is approximately 60 Mt/a. With waste decay times estimated at 10 to 20 years, there is approximately 900 Mt of active waste to account for in the stratification. Since UK landfills as a whole are too inhomogeneous a group to establish representative sites, we now divide the total into three categories or "strata" that have some common features.

The landfill sites about which most is known in the UK are the 400 or so large sites included

in the NAMGAS database (Manley 1990). This database was compiled in order to estimate the resource of exploitable gas from landfills and only includes those sites with more than 200 kt of waste in place. These sites represent the technology types that will predominate in the future so we have chosen to use them as the starting point for our stratification.

The NAMGAS sites contain an approximate total of 300 Mt of waste which represents about a third of the total active waste in the UK. In addition to the NAMGAS sites, the survey by the Water Research Centre (Reynolds 1993) suggests there are also 3000 other open sites containing less than 200 kt of waste each. If they contain on average 100 kt each, then they contain approximately 300 Mt of waste. Much of this waste is less than 10 years old and is likely to be decaying and emitting gas.

Using our initial estimate of 900 Mt of active waste suggests that there is another 300 Mt of active waste in closed sites, about which very little is known. Recent work in the US (EPA 1993) suggests that emissions from closed sites may be significant.

In the following sections, we use these approximate figures to propose a three-level stratification of UK landfills. It is not our intention to put these numbers forward as another estimate of UK methane emissions, they are used solely for devising a strategy for improving emissions estimates and for evaluating the cost-effectiveness of direct emissions measurement performed on the basis of a stratified sample.

We can now examine these strata in greater detail in order to estimate the upper and lower bounds for the emissions from each.

2.1.4 Benefits of Sampling the Sites Included in the NAMGAS Database

There is considerable uncertainty about total emissions from the NAMGAS sites. Manley (1990) estimates the methane resource recoverable from these sites to be about 1.7 Mt/a. This figure has been derived by combining calculations of the total methane potential with the results from pumping trials. As such it takes no account of oxidation effects and is therefore a reasonable upper bound for emissions from these sites.

In order to establish a lower bound for these emissions we take an optimistic view of the effects of oxidation. Estimates of the effects of oxidation reduce source-based calculations by between 10 and 50%. Hence a lower bound for emissions can be taken as 50% of the upper bound, or approximately 0.8 Mt/a. These figures suggest that for the purpose of illustrating a stratification, we can assume that NAMGAS sites are responsible for approximately 50% of UK landfill emissions and approximately 50% of the uncertainty in the estimate of the total. These figures for the NAMGAS sites were calculated using existing data and have a quality of C.

The NAMGAS sites have a degree of commonality of design and conditions across them. More importantly, they have already been categorised by hydrogeology and waste type, to a greater extent than any other group of sites. A cost-effective measurement program would measure emissions from a selection of sites in several of Manley's categories with different capping conditions. These sites would be chosen following advice from experts on the subject.

We believe that accurate direct emissions measurements performed on 20 sites over a period of one year would reduce the uncertainty in the emissions estimate for this category by approximately 50%.

As well as an improvement in the value for total emissions from NAMGAS sites, such an exercise would also provide an opportunity to compare results from current models with direct measurements of emissions.

2.1.5 Sampling Closed Sites

The second stratum in our analysis is "closed sites". Although the location and contents of closed landfill sites are not well known, it is possible to estimate upper and lower bounds for the emissions from them. Taking a liberal estimate for historic waste arisings and a long decay times, emissions from these sites could be as high as 3.8 Mt/a. In order to estimate the lower bound, we take a conservative estimate for historic waste arisings and assume that decay times are very short, this gives a lower bound of 0.2 Mt/a. These figures suggest that for the purpose of illustrating a stratification, it is reasonable to assume that closed sites are responsible for approximately 25% of UK landfill emissions and approximately 25% of the uncertainty in the estimate of the total.

The sources of the large spread in these estimates are the uncertainties in the decay lifetime and in historic waste arisings data. It gives rise to a highly asymmetric distribution of estimates with a data quality of D. The decay lifetime of degradable waste in a landfill has been investigated by various methods (Pacey 1990), but there is still doubt as to the representativeness of the data produced. The very asymmetric shape of the estimated distribution for this category suggests that a relatively simple series of measurements could be performed in order to put an upper limit on the emissions from these sites. Measurements of this kind will give a greater understanding of the time periods over which waste decays and will give greater confidence in the age at which it can be assumed that emissions are negligible. This will also help in the prediction of future emissions from landfills, and will form a useful contribution to the formulation of control strategies.

2.1.6 Sampling Open Sites not included in the NAMGAS Database

As discussed above, about one third of degradeable waste in landfills in the UK is contained in open sites that are not included in the NAMGAS database. Forming a direct estimate of the emissions from this stratum is extremely difficult. It is also difficult to propose how the uncertainties in the estimate for this category can be reduced by direct measurement (or by modelling). This is because it is an extremely inhomogeneous group, representing a large number of different types of landfill of different ages and containing differing quantities of waste. It is possible that there are further useful strata into which this category could be divided but none are evident from the data available to us. This issue requires further investigation which is beyond the scope of this report but could be usefully accomplished by extending the scope of a database of the NAMGAS type. Without further information about the sites in this category it will be difficult to propose any method for reducing the uncertainty associated with their emission estimate.

We believe that the problem of reducing the uncertainty associated with this category should be reconsidered after the results from the studies of NAMGAS sites proposed above have been obtained.

Table 22 Example of Possible Stratification of UK Landfill Sites

The numbers in this table are solely intended to illustrate the principle of stratification. They are not intended to constitute a new estimate of the total.

Stratum	Number of Sites	Waste in Place [Mt]	Fraction of Total Emissions %	Fraction of Total Uncertainty %	Data Quality
NAMGAS	400	300	50	50	C
Open non-NAMGAS	3000	300	25	25	E-
Closed	8600	300	25	25	D
Total	12000	900	100	100	D

2.1.7 Measurement Details and Costings

Throughout the above discussion it has been assumed that it is possible, with the techniques described in Part II section 2, to obtain a reliable estimate of the total annual emission from a given landfill site. Costing requires a more detailed look at exactly how the measurements could be performed.

The optical integrated-path monitor described in section 2 of Part I is the most cost-effective way of measuring total methane flux at any point in time. It has the immediate advantage that by measuring over a long path it avoids the problems resulting from the spatial inhomogeneities of landfill emissions. Further, it has been shown that it can give accuracies of up to 15% for instantaneous emission figures in appropriate meteorological conditions. However, these accurate total flux measurements require the light path to be raised up to 15 m above the ground by a system of mirrors on scaffolding towers or masts. This can make unattended operation over extended periods difficult. It is therefore proposed to combine flux measurements using elevated light paths with longer-term monitoring at ground level with either flux boxes or optical integrated-path monitors. These long-term ground-level measurements will provide an average over the diurnal, seasonal and meteorological variations in emissions, which are the major source of the representativeness problem associated with total flux measurements made over a short period of time.

Taking all these factors into account, and assuming a statistically valid sample of between 15 and 20 sites, the measurement programme (without any associated emission modelling) described above would cost approximately £180k. This total could be split over an 18-month period.

Another advantage of this approach is that it can be performed in collaboration with emission modellers who can then use the results to improve and verify their calculations. The sites will be chosen in consultation with industry experts in order to make the most of this opportunity.

2.1.8 Conclusions - Landfill

We have presented an analysis of the uncertainties in the current estimate of methane emissions from landfill and possible means of reducing them. There is a need for a focused programme of direct emission measurements. This would both reduce the uncertainty directly and also improve and verify existing calculations. It is important that such a programme is effectively coordinated with existing activities in this area. A strategy could be based on the following elements.

(i) The NAMGAS database provides the best data on methane emissions from large UK landfills representative of the technology that will be used in the future. We believe that the emissions from these sites are currently approximately 50% of the UK total for landfill. A series of targeted measurements costing up to £180k would reduce the uncertainty associated with these sites by about a half. Since these sites are responsible for approximately 50% of the uncertainty due to landfill emissions, we believe that such a series of measurements could reduce the total landfill uncertainty by up to 25%. Such measurements would also enable the NAMGAS sites to be used for a valuable comparison with the emissions calculated from models.

(ii) There is great uncertainty over the quantity of waste in closed sites and the emissions from these sites. We have estimated that the emissions from these sites are approximately 25% of UK landfill emissions. This uncertainty could be reduced considerably by a series of measurements targeted at establishing the upper limit for emissions from sites of different ages (10, 15, 20 years). Such a series of measurements would cost approximately £40k and reduce the uncertainty associated with these sites by at least a half, with a consequent reduction in the uncertainty associated with landfill emissions by between 15 and 20%. This exercise would yield a lot of information about the decay rate of methane missions which is needed in order to predict future methane emissions.

Using data available at present, not much can be done about active sites that are not included in the NAMGAS database because they are an extremely inhomogeneous group. An extension of the methodology described in (i) could be considered after its results have been evaluated.

There is an important continuing requirement to update existing models with data from current programmes of all types in this area. Some coordination will be necessary to ensure that this takes into account the need for accurate emissions calculations. This coordinating role could be effectively played by a small working group representing each of the organisations with expertise and interests in this area.

The work proposed above will contribute to increasing the accuracy with which the UK methane inventory is calculated, and hence should produce an "iterative" improvement to the overall figure.

2.2 ANIMAL RESPIRATION

It is clear from Part I of this report that this sector is the major source of uncertainty in the inventory after landfill. This is particularly true because it is believed that the uncertainty figures, (which were taken from the Watt Committee), understate the true level of uncertainty.

As discussed in Part I, the heart of the problem is whether the emission factors used are truly representative of the situation in the UK. It has been shown (Milton 1992) that optical integrated-path techniques can produce estimates of methane production by cattle (which accounts for 75% of the methane in this sector). These estimates are made in the field, without the problems associated with disturbing the animals. There is, however a need to collate existing data in order to specify an appropriate methodology for improving this estimate. Problems that need to be addressed include the effects of different feeds, grasslands, animal breeds and husbandry practices.

2.3 ANIMAL WASTES

After animal respiration, the next largest uncertainty in the inventory is that due to emissions from animal wastes. The major source of uncertainty in this sector is the methane conversion factor (defined as the proportion of potential methane that is actually generated from the waste). There are also questions that should be addressed by a data collation exercise, such as the validity of the data on the quantity of waste produced and the details of the treatment process, but the biggest reduction in uncertainty would come from a programme of measurements aimed at the methane conversion factor. The precise details of such a programme would need to be worked out in collaboration with relevant experts. A reduction in the uncertainty of some tens of percent could be expected.

2.4 COAL MINING

This is a sector with emissions that are responsible for only a small fraction of the national total. The data quality is high as the estimates are based on a large number of direct measurements. An improvement could be achieved by monitoring all mines continuously, but the expense involved would not be justified by the relatively small improvement to be gained. The uncertain future of the industry is a further reason for not proposing work to improve the estimates in this sector.

2.5 GAS LEAKAGE

In general, this is an area which has been well covered by the Watt committee. By far the largest source of emission is the low pressure distribution network, which has been estimated by a large number of direct measurements. An area that merits further investigation is domestic losses after the meter for which present estimates are based on a very small number of measurements. A small laboratory-based programme of experimental measurements on typical domestic gas components and systems would help to reduce this uncertainty.

2.6 SEWAGE

The total estimate for this sector has a large uncertainty which arises from a large number of factors. The most promising method of improving the estimate from this sector would be measurements on sewage lagoons and possibly anaerobic digesters. It is not possible to suggest the details of such a campaign without further investigation of the representativeness problem within the sewage sector. Accordingly it is hard to quantify the likely costs and benefits of such a programme, but it would be likely to cost over £20k and reduce the uncertainty by approximately 20%. It should be borne in mind that this sector is a relatively minor contributor to the UK methane budget. There is also a requirement (as discussed in Part I), for improved data on waste arisings and quantities of sewage treated by different methods.

2.7 OFFSHORE OIL AND GAS

This is an area where the emission estimate has fallen dramatically recently, due to a completely new set of figures produced by the industry. Although measurements are technically difficult in this sector, there is a need for a detailed critique of existing estimates. In particular, an investigation is justified into the apparent discrepancy between the figures for total VOC emissions for this sector and those for methane, which suggest that the figure for methane might be larger than stated.

2.8 STATIONARY COMBUSTION

In this sector there are major uncertainties from the emission factors for the domestic combustion of solid fuels and natural gas. These have been taken from a limited number of measurements and have a wide spread and low data quality. There is therefore a need for improved measurements of emissions from these sources. This is an area which is likely to be most profitably tackled by a programme of laboratory measurements.

2.9 SUMMARY

This section describes in detail how direct emission measurements can be applied cost-effectively to the improvement of methane emissions estimates. Two complementary measurement programmes have been proposed for the reduction in the uncertainty in landfill emission estimates. Both of these programmes would lead to a significant reduction in the uncertainty of total UK methane emissions. The application of direct measurements to seven other sectors of the inventory has also been discussed. Each of these would result in a reduction in the associated uncertainties, but would make a much smaller impact on the UK total than the measurements of landfill emissions. The proposals are summarised in Table 24 in the Conclusions.

Part III The Application of Measurements to the Verification of the Estimate of UK Methane Emissions

The purpose of this chapter is to outline various measurement and assessment programmes that would improve the completeness and accuracy of the National Atmospheric Emissions Inventory (NAEI) for methane and provide information regarding unidentified or unquantified sources. Two programmes are proposed. The first is an assessment of available information regarding methane sources that are not presently included in the NAEI. The second programme uses ambient concentration measurements together with inverted dispersion modelling to verify the emissions estimates calculated from the inventory.

1 POSSIBLE SOURCES OF METHANE OMITTED FROM THE UK EMISSIONS ESTIMATE

Work on Part I of this study has identified a number of potential sources of methane emission that are not presently included in the National Atmospheric Emissions Inventory (NAEI):

The chemical industry (other than fuel combustion),
Secondary fuel manufacture,
Crude oil distribution,
Bitumen refining and use
VOC abatement devices,
"Semi-natural" sources.

1.1 The chemical industry (other than fuel combustion)

An approximate estimate of national methane emissions from the chemical industry was derived in Part I, using data reported by one company. In order to improve this estimate it would be necessary to compile a list of methane emitting processes, establish suitable emission factors for each process and quantify the corresponding process statistics. A number of methane emitting processes carried out by the chemical industry were identified in Part I. Further processes could be identified by literature review and through discussions with the industry via the Chemical Industries Association and its member companies.

Methane emission factors could possibly be estimated for some processes by using the mass balance principle. However, for most processes a better estimate could be made by direct measurement of stack or fugitive emissions as appropriate.

1.2 Secondary fuel manufacture

The manufacture of secondary fuel includes solid smokeless fuels and coke from coal (which involves the partial pyrolysis of coal which drives off volatile components). It is likely that

these processes release methane as well as other organics. This process has been identified in the NAEI as a source, but with zero emissions. As with the chemical industry, direct emissions measurements and the collection of process statistics would allow a better estimate to be made.

1.3 Crude oil distribution

There are problems of definition over crude oil distribution, and it is not clear whether methane emission estimates for this source have been included within estimates for other sources. The problem arises in part because of uncertain industrial representation: UKOOA represents oil exploration and production activities, whilst UKPIA represents refinery activities. Within these activities, emission sources may be present which have not, to date, been clearly identified. In particular, crude oil can be held in stabilising tanks prior to delivery to the refinery where methane and other very light fractions can be lost. As a first step, it would be necessary to clarify source coverage from UKOOA's and UKPIA's emission estimates. If it is the case that some sources have been omitted, then emission estimates could be obtained by measurement coupled with throughput statistics.

1.4 Bitumen refining and use

Bitumen refining is carried out partly by integrated petroleum refineries and partly by specialist bitumen refiners. Bitumen is produced from some crude oils (notably Venezuelan) by vacuum distillation but for some commercial production from medium crudes, it is stiffened by blowing with air. This process produces "foul gas" which is likely to contain some methane although most is thought to be incinerated. The steps needed to establish a first order estimate of emissions are similar to those described for secondary fuel manufacture.

In the UK, about 80% of bitumen is used in road construction and repair (1350 kt in 1988). The bulk of this is "straight-run" bitumen with a volatile content in the region 1 to 2%. Of the remaining 20%, most is used for roofing. The handling and application of such bitumen and bitumen-containing products can be a source of methane emissions. Tyler *et al.* (1990) undertook limited measurements of methane emissions from bitumen pavements and found that the rate of release was dependent on sunlight intensity and observed methane emissions from pavements more than two years old.

1.5 VOC abatement

VOC abatement devices can emit methane as a product of incomplete oxidation of VOC's, these include: thermal and catalytic incinerators, biofilters , bioscrubbers, and flares. With the exception of flares, which are best treated as sub-components of the oil and chemical industries and landfills, such abatement devices are not likely to be major sources of methane emission.

1.6 "Semi-natural" Sources

Man's activities in land management, agriculture and waste disposal (other than landfill) may have increased natural methane emissions or stimulated additional emissions in a number of ways.

Peat extraction is not widely carried out in the U.K., but disturbance of peatlands can result in methane emission (Lacroix 1993) so that any development of such areas, e.g. in planting for forestry, may result in emissions. Methane is a component of groundwaters and Coleman *et al.* (1988) have shown that methane may be released to the atmosphere from extraction processes. Mineral extraction (other than coal) has also been shown to release methane, although the amounts involved are very uncertain (Lacroix 1993). Shallow submarine sediments have been shown to be a source of methane from the bacterial degradation of organic materials (Hovland *et al.* 1993). Whilst this may be considered a "natural" process, the enhancement of the organic content of coastal waters from sewage outfalls and disposal should be considered as essentially anthropogenic. Marlowe *et al.* (1992) have estimated that the emission of marine and inland waters, mudflats and soils are of the order of 360 kt/a.

Whilst these sources are considered here as making only a relatively small contributions to total UK methane emissions, they should not be discounted, as the sum of the parts may amount to a significant contribution, as has similarly been shown in the estimation of ammonia emissions from the U.K. (Lee and Dollard 1993).

2 METHODOLOGIES FOR VERIFYING REGIONAL AND NATIONAL INVENTORIES

There is a considerable amount of work being carried out throughout the world on the generation of methodologies for verifying inventory estimates (Saegar 1992). This section sets out the components of a programme that uses ambient measurements to improve estimates of anthropogenic emissions. A programme would include a paper assessment of the source strength of a selected area followed by measurements of environmental concentrations carried out in a manner that would allow the source strength to be estimated by modelling. Agreement between the two estimates would give confidence that the prior knowledge of emissions was correct, and that no significant sources had been omitted. Such a programme would have to take account of any natural emissions or sinks in the chosen source area. If the technique were applied in an urban area, or an area of intensive livestock production, the source inventory that would be validated would be dominated by anthropogenic sources. However, if it were used on an area as large as the UK, the inventory would also have to account for natural sources.

2.1 Definition of specific regional inventories

An essential preliminary would be an assessment of the emissions inventory from areas

selected for the verification measurements. This activity would include a cataloguing of all emission sources in the selected areas, estimation of emissions of each source type and a quantification of the uncertainty in the resulting total emission. Information on the geographical distribution of sources within the selected areas would also be assembled. This procedure would be carried out for candidate urban, rural and industrial areas. The resulting emissions estimate would be compared with the results of measurements in order to verify the emission factors used in the NAEI. The results would also aid selection of sites for sampling and confirm the suitability of selected areas for the measurement campaign.

2.2 Assessment of available data

There are a limited number of data sets of ambient methane measurements. Many of the published measurements are for remote measurement sites and provide information only on the global methane budget. Two data sets relating to the UK have been identified.

Simmonds and Derwent (1991) examined a few months of methane data from Mace Head in order to assign regional contributions according to 45° sectors, assuming that the same emission rate per head of population applied to all areas of Europe. On this basis they were able to estimate the total methane emissions from Europe, including the UK. Evidence was found for an additional source in the north-easterly sector which may have its origins in emissions from tundra and wetlands. The simple formulation of the model (straight line trajectories, constant immersion height, background gas concentrations at the start of the trajectory and surrogate emission statistics) may cause significant uncertainty in the source estimate. Nevertheless, this approach showed how such observations could be put to good use. There would be value in updating the evaluation of the data to examine trends and seasonal fluctuations. It would also be worth examining the data in more detail to determine whether better spatial resolution of sources could be achieved.

Measurements of methane, amongst other trace gases, made at Imperial College in central London were recently reported by Goldstone *et al.* (1992). These data (possibly in combination with the Mace Head data) could be applied to estimate the methane source strength in London. Such a study would also provide an opportunity to compare methods for interpreting urban data (described in the following section).

2.3 Experimental Verification of Emissions Estimates for an Urban Area

Methane emissions in a major urban area are likely to be totally dominated by artificial sources. Landfill sites, often around the periphery, are expected to make the greatest contribution, but all of the domestic and industrial activities will be represented to some degree, and any substantial omission or error in the inventory of man made sources should be detectable if the emissions from a large city are determined from measurements at a suitable level of accuracy.

In section 2 of Part II, various possibilities were considered. For instance, measurements made from an aircraft downwind of a large city, similar to those of Blake *et al.* (1993), would give a measure of the methane emission during a brief period, and would also give some information on the homogeneity of the emission field. Ground-based measurements using

vehicle-borne point measuring instruments or, preferably, DIAL equipment mounted on a vehicle, could probably achieve the same result at lower cost, although the possibility of the results being unduly influenced by local sources, close to the point of measurement, would need vigilance. Either measurement method would yield only short-term data, and variability in sources over periods from hours to annual cycles would complicate interpretation.

Fixed point measurements over a period of about a year would overcome the brevity of record from a mobile measurement campaign, and would allow the annual average emission from the city to be estimated. Location of measurement points within the city would maximise the signal, and, taking advantage of variations in wind direction, would provide some information on the spatial distribution of sources. However, locating the measurements within the city would require the issue of representativeness to be addressed.

Alternatively, measurements could be made at a distance of one or two diameters outside the city. Such measurements are unlikely to be excessively sensitive to the distribution of sources within the urban area. Periods when a measurement point was down-wind of the source area would provide information on the emission strength. Other periods would give a measure of the appropriate background.

The selection of the city or town for the urban area validation study is an important question. A verification study based on London would confirm a large fraction of the national emission, but the complexity of the large source area would cause difficulty in interpreting any difference between the observed and expected data. A smaller and simpler town would result in easier resolution of such a discrepancy, but the emission factors applying to such a location might reflect local conditions, and would be representative of national conditions only if the location were chosen with care.

The isotopic composition of methane can be used in studies of sources to good effect. Since ^{14}C is present in fossil fuel related emissions, the concentrations of ^{14}C in atmospheric methane gives information on the fraction of the methane that derives from modern biogenic, rather than fossil, sources. Source attributions can also be determined using variations in the stable $^{13}C/^{12}C$ isotope ratios. Such a study was undertaken by Thom *et al.* (1993) who determined the contribution of various sources for an area within a radius of 500 km of Heidelberg. Such measurements could provide valuable additional information in the context of urban measurement campaigns, taking samples at intervals for isotopic composition analyses.

2.4 Regional Rural Emissions

Some areas in the west of the UK are used intensively for cattle production and contribute a substantial fraction of the emissions attributed to this activity. Emissions occur from the animals themselves and from associated processes such as grazed fields, slurry pits and dung heaps. Thus, the emissions are expected to be widespread over the area of several counties. Measurements of ambient methane concentrations within such an area would provide an opportunity to verify the overall emission factors for livestock. The use of a year-long sequence of measurements would ensure that seasonal and shorter term fluctuations were adequately represented.

The measurements would allow the source strength in several wind direction sectors to be estimated. Verification of the emissions inventory could then be achieved by comparing estimates with values derived from calculations of source emission rates.

2.5 National Verification of Sources

Three different approaches are considered for verifying national emissions estimates. The first uses an aircraft to make measurements around the coast of the UK. The principal disadvantage of such a campaign is the cost involved. Furthermore, the representativeness of the measurements to verify a national flux on an annual basis must be carefully considered. For example, in two such recent flights across high elevation natural ecosystems in Scotland, contrasting results were obtained because of differences in environmental conditions. (Derwent, personal communication). Spatial variability in the concentration in the plume from the UK may provide valuable information on source regions within the country.

A second approach would be to establish a sparse monitoring network of fixed-point monitors across the UK for a period of one year or so. This is essentially an expansion of one of the approaches suggested for regional inventory verifications. There are several advantages to this; there are established networks already running across the UK in various urban and rural locations as part of the Enhanced Urban Network and the Rural Monitoring Network, existing maintenance programmes at a limited number of sites could be adapted to include automated methane measurements, and telemetry systems for data gathering are already in place. The data bases that result would include information on the spatial distribution and temporal variability of emissions that would aid identification of unexpected sources.

A further option is to locate a monitoring station in the North Sea or on the west coast of the European mainland. Information on the total source strength and the distribution of sources in the U.K. would be collected in west winds, whilst information of European emissions and emissions from the North Sea oil and gas fields would result from other wind directions.

3 SUMMARY

Programmes of paper evaluation and measurements carried out to improve and verify the national emissions inventory, would make the inventory more comprehensive and could indicate the presence of any substantial unidentified sources. Six types of source have been described that are not treated thoroughly in the existing estimate of UK methane emissions. A programme should be targeted at evaluating these source categories on the basis of information available from industry and the literature. Such a study would increase confidence that the National Atmospheric Emissions Inventory is comprehensive, with no substantial omissions.

As has been shown, a wide variety of approaches can be used to verify regional and national emission estimate including both measurement and modelling techniques. For a convenient comparison, they are summarised in Table 23.

Table 23. Summary of options for unquantified sources and inventory verification.

Activity	Benefit	Cost
Unquantified Sources		
Identification of methane emitting processes requiring measurement in the chemical industry.	Improved confidence and greater transparency of emission estimates	
Study of emissions from secondary fuel manufacture.		
Investigation of possible omitted sources in the oil production and refining chain.		£20k
Approximate quantification of emissions from bitumen use.		
Study of possible large emissions from 'semi-natural' sources.	More complete inventory of 'background' sources.	
Assessment of Pre-Existing data		
Modelling of London and Mace Head data sets.	1.Estimation of source strength for London 2.Trends in European (and UK?) source strength. 3.Experience in interpretation techniques.	£30k, assuming free access to datasets.
Urban regional		
City and regional inventories.	1.Basis of assessment of verification measurement. 2.Aid selection of measurement sites.	£50k
Point location in a city	1.Verification of long-term city emission inventory. 2.Distribution of sources within city. 3. Seasonal variation.	£50k assuming siting available
Two point locations near a city		£90k assuming siting available
LIDAR city plume measurement.	1. Verification of short term source inventory of a city.	£30k per week
Aircraft city plume measurement		£100k
Rural regional		
Point measurement (one site in an agricultural area).	1.Verification of long-term agricultural emission. 2.Seasonal variability.	£50k assuming siting available
National		
Aircraft measurement off E. Coast	1.Short term verification of national inventory. 2.Some information on source areas.	£100k
Onshore network of monitors.	1.Verification of national emissions inventory. 2.Information on source areas and continental emissions.	£50k per site
Fixed monitor site in N.Sea.		£100k

The potential for verification studies has been discussed on urban, rural and national scales. Data available from Mace Head and Imperial College, London, provide an opportunity to carry out a pilot investigation of the use of ambient measurements to assess long-term mean inventories, and also to use variations related to wind direction, time of day and season to seek information on individual source categories. In the case of urban and rural studies using point measurements, it is anticipated that data acquired from such studies would be used in combination with available meteorological information and remote background data from Mace Head on the west coast of Ireland for comparative purposes.

The data available from Mace Head and Imperial College are not suitable for verifying regional or national emissions which could, however, be verified by a separate field programme. A limited number of samplers would be operated at sites selected to provide information regarding a large fraction of the inventory. Simultaneous operation would be advantageous, since interpretation of the results would be facilitated in some areas.

Short-term measurements using aircraft have the advantage of providing spatial resolution. The full costs are high in relation to the time scale covered and the data from individual measurement campaigns may not be representative of long-term emissions. These measurements would contribute to the evaluation of the national emission rate and would also provide some information on the distribution of methane sources. Additional source resolution could be achieved using isotope ratio measurements. Both the radioactive isotope ^{14}C and the stable isotopes ^{12}C, ^{13}C, ^{1}H and ^{2}H, carry information about the origins of methane. Accordingly, it may be necessary to arrange for the collection of additional samples during aircraft or point-sampling campaigns, after the concentration measurements had been assessed.

Conclusions and Summary of Recommendations

The UK emissions inventory for methane was reviewed and recalculated in Part I of this report. For the first time there has been an explicit analysis of the uncertainties in each part of the calculation which has led to the identification of many areas of the inventory where there is a requirement for new data. Some of these requirements can be satisfied by the use of the direct emission measurement techniques described in Part II, while others require a coordinated approach of continued modelling, data collection and measurement. These issues have been addressed, sector by sector, in Part II.

In addition to the need for improvements to sector estimates, there would also be some benefit in considering how the methodologies described in Part III could be used to verify the methane emissions inventory as a whole (either on a local, regional or national basis). This could be done by measuring ambient concentrations arising from the whole UK or from a region for which the inventory could be recalculated specifically and relating these measurements back to source strengths by the use of plume dispersion modelling techniques.

1 COORDINATION AND PRIORITISATION

As the first part of this report illustrates, there are a large number of organisations involved in collecting the data used for estimating UK methane emissions. At present, most of this data is not acquired or compiled with the primary objective of estimating emissions. Consequently, it does not always have the scope or accuracy that is required for this application. The diversity of organisations with significant expertise in this area, and the range of the options available for making improvements mean that there is a significant task of coordinating any future programme aimed at improving emissions estimates. We believe that there would be great benefit in the establishment of a small coordination group with representatives from UK organisations with expertise and interests in methane emissions. This group could undertake the following coordinating tasks:

- Collating information originally derived for other reasons and making a critical assessment of its applicability to estimating emissions.

- Providing expert advice on exactly how measurements should be targeted for best cost-effectiveness.

- Providing expert interpretation of measurements and feeding this back in the form of adjustments to the ongoing measurement program, or improvements to existing models.

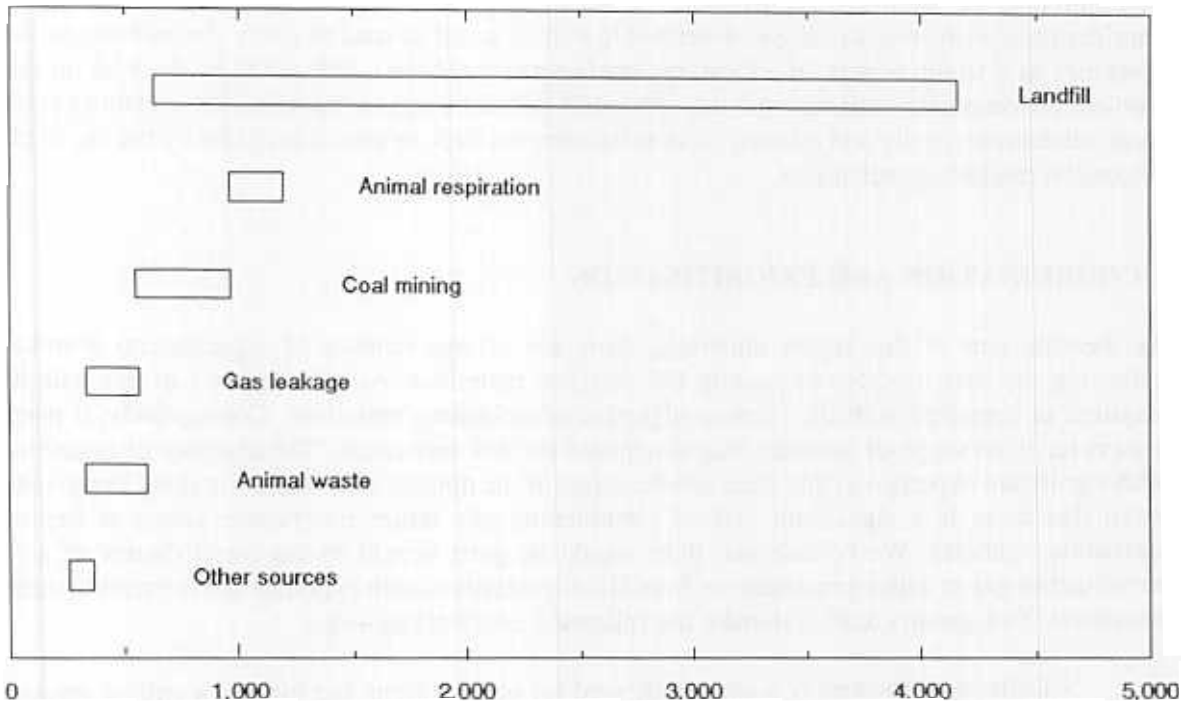


Figure 11. Range of estimated methane emissions by source

2 SPECIFIC RECOMMENDATIONS FOR IMPROVING SECTOR ESTIMATES

The uncertainty in the estimated emission for each of the sectors used in the NAEI is shown in Figure 11. The extent to which the uncertainties in each sector could be reduced by the use of direct measurement techniques is shown in Table 24.

It is clear from the data presented in this report that the outstanding requirement for improving the UK methane emissions estimate is for an improvement to the estimate of landfill emissions. Present estimates are based on models that use relatively few parameters directly determined from in situ measurements. The use of direct emission measurements would allow these estimates to be improved. We have concluded that this could be done most cost-effectively by the use of several measurement techniques together with a stratified sampling methodology. This approach would result in a significant reduction in present uncertainties. A programme of measurements focused on the largest sites and costing approximately £180k will result in a direct reduction of 25% in the uncertainty estimate of landfill emissions. In addition to this direct reduction, a programme effectively coordinated with other activities in this area, would also lead to a better understanding of the processes involved, and hence a further reduction in the overall uncertainty.

The application of these techniques to other sectors has also been considered. In each case an improvement in the emissions estimate could be achieved, but the overall impact on the total UK uncertainty is considerably smaller than using similar resources on the landfill sector. Table 24 contains a summary of these sector-specific recommendations.

3 INVENTORY VERIFICATION

Two principal activities in the area of inventory verification were described in Part III of this report. The first is an analysis of those sectors of the current inventory from which it is believed that some sources have been omitted. These have been summarised in Table 23, along with some proposals for correcting the omissions.

The second activity is the verification of the inventory on a regional or national scale by the comparison of calculated emissions with measured ambient concentrations. A range of proposals has been presented and are costed in Table 23. These techniques have been applied for other substances, but it is difficult to predict the accuracy achievable for methane which has a heterogeneous and uncertain source distribution. However, there is considerable international interest in the verification of inventories using these techniques which are entirely complementary to the direct approaches to inventory improvement described elsewhere in the report.

Table 24 Summary of Recommendations

Sector	Recommendation	Cost £k	Potential Improvement to sector uncertainty	Potential Improvement to sector data quality
Landfill	Measurement programme based on NAMGAS sites	180	25%	D to C
	Measurement programme based on closed sites	40	15 - 20%	No change
Animal Respiration	Measurements of emissions from cattle under a variety of conditions	?	50%	D to C
Animal Wastes	Measure methane conversion factors	10 - 40	30%	D to C
Coal Mining	No proposal	-	-	-
Gas Industry (onshore)	Investigation of "post-meter" losses	30	5% ?	No change
Sewage	Measurement of sewage lagoons	20 - 80	20 - 50%	No change
Offshore Oil and Gas Industry	Critique of industry's estimates	4	?	No change
Stationary Combustion	Measurements of emissions from domestic combustion of solid fuels and gas	25 - 40	5%	No change
All sectors	Coordination group	20 pa	-	-
Unquantified sources	Assessment of emissions from sources omitted from present inventory.	20	-	-
Verification	Described in detail in Table 23			

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