The Potential for Methane Emissions from Groundwaters of the UK

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ABSTRACT

Methane (CH_4) is only a trace constituent of the atmosphere but an important greenhouse gas. Although groundwater is unlikely to be a major source of atmospheric CH_4, its contribution to the CH_4 budget of the UK has up to now been poorly characterised. Groundwater CH_4 concentrations have been measured on 85 samples from water-supply boreholes and a further eight from other miscellaneous water sources. Concentrations in abstracted groundwaters ranged from <0.05 – 42.9 µg/l for Chalk, <0.05 – 22 µg/l for the Lower Greensand, 0.05 – 21.2 µg/l for the Lincolnshire Limestone and from <0.05 – 465 µg/l for the Permo-Triassic sandstone. Having the largest abstraction volume, the Chalk is likely to be the main UK groundwater contributor to global CH_4 emissions. A calculation to estimate the total emissions of CH_4 from water supply groundwater sources based on the median and the maximum CH_4 concentrations gave a values of 2.2×10^{-6} Tg/yr and 3.3×10^{-4} Tg/yr. Estimates show groundwater contributes a maximum of 0.05% of all UK CH_4 emissions and a further two orders of magnitude less in terms of the global CH_4 budget. Other groundwater sources such as inflows to tunnels may have significantly higher CH_4 concentrations, but the volume of water discharged is much lower and the overall amount of CH_4 outgassed is likely to be of the same order as the aquifer release. The generally low concentrations of CH_4 in groundwater supplies suggest no threat of explosion, although groundwater released by excavations remains a hazard.

Key Words: Methane (CH_4), Groundwater, Emissions, UK, Explosion hazard.

INTRODUCTION

Methane (CH_4) has both general and local impacts on the environment. On the one hand it is a greenhouse gas, some 20 times more potent than CO_2 (US EPA, 2002), while on the other it poses a potential explosive threat when build-up occurs in confined spaces (Hooker and Bannon, 1993).

The role played by groundwater in both the above areas has been poorly understood for the UK. Admittedly it has never seemed likely that CH_4 originating in, or transported by, groundwater would be a major source of greenhouse CH_4 or a widespread explosion hazard, yet information on baseline concentrations has not hitherto been available to confirm this. The present study examines CH_4 concentration data from three types of source (major water-supply aquifers, igneous and metamorphic rocks, and high-methane waters) to characterise the groundwater CH_4 inventory rather better. To do this 93 samples have been analysed and interpreted. The potential contribution of groundwater CH_4 to the global carbon budget has been assessed.

BACKGROUND

Origins of methane

Methane is a common trace component in groundwater and occasionally constitutes a major carbon pool (Barker and Fritz, 1981; Cramer et al., 1999; Geyh and Künzl, 1981; Parkin and Simpkins, 1995; Watanabe et al., 1994). Methane of varying origins can accumulate in near-surface groundwaters: abiogenic CH_4 has been identified in groundwater in the Canadian Shield (Sherwood et al., 1988); in ophiolite terrains (Neal and Stanger, 1983); and in mid-ocean ridges (Welhan, 1988). Thermogenic CH_4 (produced by thermal decomposition of organic matter) is associated with coal, oil or gas fields (Tissot and Welte, 1984), or indeed any situation where organic-rich sediments are thermally stressed (Simoneit et al., 1988). Biogenic CH_4 (bacterially-produced) has been found in marine environments (Schoell, 1988) and anaerobic groundwater environments, including peat bogs, lignite deposits, and glacial, lacustrine and aeolian sediment (Barker and Fritz, 1981; Coleman et al., 1988; Grossman et al., 1989). The concentration of CH_4 in natural waters can range from <1 – 15,000 µg/l which reflects its variation in source from anaerobic environments (Thurman, 1985).

Abiogenic CH_4 is not an issue for the UK; thermogenic CH_4 may be a local factor, but the most widespread production of the CH_4 found in groundwater is likely to be biogenic.

Methanogenesis

Methanogenesis is a complex process and can proceed via several possible metabolic pathways and use several possible substrates. The two major metabolic pathways are acetate fermentation,

$$CH_3COOH \rightarrow CH_4 + CO_2 \qquad (1)$$

and CO_2 reduction,

$$CO_2 + 4H_2 \rightarrow 2H_2O + CH_4 \qquad (2)$$

Methanogenesis represents the last of a series of reactions where electron acceptors such as O_2, NO_3^-, Mn^{4+}, Fe^{3+} and SO_4^{2-} are used sequentially to oxidise organic matter. Conditions most favourable to biogenic CH_4 formation are found in anaerobic and low Eh environments, such as confined groundwaters (Bishop and Lloyd, 1990), groundwaters from beneath rapidly developing cities (Lawrence et al., 2000) and, in particular, leachates beneath landfill sites (Christensen et al., 2001) and organic waste stores (Gooddy et al., 2002) where a usable substrate exists to provide energy. However, under the well-oxygenated conditions typical of unconfined aquifers, methanogenesis would not be expected to be a major process.

Groundwaters in the UK

The habitats of groundwater in the UK vary considerably, from low-permeability mudrocks to the minimal porosities of crystalline (igneous and metamorphic) rocks, and ultimately the relatively high porosities of the water supply aquifers of (mainly) the south and east. Each of these habitats has different implications for CH_4 production and ultimately emissions to the atmosphere.

Main aquifers

Groundwater meets over 30% of the water demand in England and Wales (DETR, 1997), 8% in Northern Ireland and 5% in Scotland (Bell et al., 1997). The regional differences reflect the distribution of aquifers and the more favourable geological conditions for surface water resource development in the most northern parts of the UK. Four aquifers - the Chalk, Permo-

Triassic sandstone, Lower Greensand and Lincolnshire Limestone - provide almost all groundwater in the UK (see Table 1) accounting for 95% of abstractions (Downing, 1993).

The Chalk aquifer is the most important in the UK since it accounts for more than half of the groundwater used. It outcrops largely in the south-east of England where population density is high, rainfall is low and there are few sites for suitable surface reservoirs. The Lower Greensand also forms an important aquifer in south-east England. Despite the relatively small outcrop area of the aquifer, the high storage and generally good quality of water render it a reliable source of water for domestic and industrial use. The Permo-Triassic sandstones form the second most important aquifer in the UK, supplying around a quarter of licensed groundwater abstractions. The aquifer provides important groundwater resources, especially in central and northern England where a number of large towns depend on it for their water supplies. The Lincolnshire Limestone is important only in eastern England but is the most hydrogeologically significant part of the Jurassic limestones.

In each of these aquifers redox conditions vary depending on the degree of confinement of the aquifer, with the extent of transition zones depending critically on the degree to which local flow systems are developed and the nature of the aquifer: for example, the well-developed fracture porosity of the Chalk and Lincolnshire Limestone promotes a greater degree of mixing between waters than is found in the sandstone aquifers (Darling et al., 1997).

When evaluating CH_4 emissions from these aquifers, consideration must be given to their discharge characteristics. In their natural state, prior to perhaps 150 years ago, any discharge from aquifers would have occurred via springs, or baseflow to rivers, or beneath the sea. Since then, the widespread development of supplies based on groundwater has distorted the natural state to an unknown extent. Many uncertainties remain regarding the total amount of discharge; the only semi-reliable data relate to the amount of groundwater abstracted.

Crystalline rocks

The so-called ‘hard rocks’ do not hold extensive groundwater resources and are generally used only for local domestic water supplies. Relatively high concentrations of CH_4 have occasionally been recorded from groundwaters of shield areas such as Canada and Sweden, but the CH_4 is most likely to have come from overlying Quaternary drift (Barker and Fritz, 1981). The possibility of ‘deep’ CH_4 emanating from crystalline rocks (Gold, 1998) has been considered in some detail but has been ruled out for the UK (Bath et al., 1986). The hard-rock data considered here were obtained from inflows to hydro-electric tunnels in the Scottish Highlands. Most of these inflows are at a considerable depth below the land surface and therefore should contain representative concentrations of any CH_4 produced *in situ* or otherwise acquired.

Mudrocks

Owing to their low permeability, the natural discharge of groundwaters from mudrocks is likely to be a very small proportion of the UK total. The generally poor quality of such groundwaters means they are rarely abstracted for water supply purposes. However, such rocks are sometimes tunnelled for water transfer between catchments, either for water supply or hydroelectric purposes, or for more general civil engineering reasons. The often high organic content of mudrocks, together with the slow rate of water movement through them, promotes the development of elevated dissolved CH_4 concentrations. Only when these waters are artificially released by tunnelling or other excavation can they be recognised.

Solubility and explosion hazard

Methane is a moderately soluble gas (Wilhelm et al., 1977). Under a typical atmospheric concentration of 1.75 parts per million by volume (IPCC, 2001), the amount of CH_4 dissolved in water at 10°C (typical UK recharge temperature) is some 7.9×10^{-6} cc (STP) per litre at equilibrium. This is equivalent to a concentration of 0.056 µg/l.

However, if the local concentration of CH_4 within the subsurface rises above the atmospheric partial pressure of $\sim 1.75 \times 10^{-6}$ bars, the amount of dissolved CH_4 will increase proportionally. Should this water become exposed to the atmosphere, equilibrium will be re-established and CH_4 will be outgassed until the extremely low background composition is reached. From the explosion hazard point of view, the consequences of this are most significant where discharge occurs into a confined space, for example a building or excavation.

Methane becomes an explosion hazard at concentrations of 5 - 15 % by volume in air (Hooker and Bannon, 1993). Since confined spaces vary both in their size and degree of ventilation, no universal rules can be applied to what may constitute a hazardous dissolved concentration in individual cases. However, if CH_4 exceeds 5% by volume of the gases dissolved in a groundwater, and the sum of these gas partial pressures is greater than atmospheric, there is a potential for CH_4 to outgas and reach or exceed the lower explosive limit. To put this in terms of concentration, a minimum CH_4 partial pressure in excess of 0.05 bars would be required, equivalent to a value of approximately 1600 µg/l.

A more detailed consideration of CH_4 release rates, ventilation and accumulation is provided in Hooker and Bannon (1993).

SAMPLING AND ANALYSIS

The locations of sampling sites referred to in this study are shown in Figure 1. Samples of groundwater were collected over a period of time by a variety of methods, all ultimately relying on the outgassing of a known amount of water into a headspace of known volume. The partitioned headspace gas was analysed by gas chromatography using a 2 m column containing Porapaq-Q held isothermally at 25°C. Helium at a flow rate of 30 ml/min was used as a carrier gas and detection was by flame ionisation. Certified canned gas standards were run with the samples in order to calibrate the results. Ultimately a detection limit of 0.05 µg/l was achieved by using this approach.

RESULTS

Main aquifers

A statistical summary of CH_4 data for 85 groundwater samples collected from the four major aquifers in the UK is shown in Table 2. Concentrations are below 10 µg/l for 95% of Chalk samples, 92% of Permo-Triassic sandstones, 89% of Lincolnshire Limestones and 65% of the Lower Greensand. In terms of the median values, Lower Greensand > Lincolnshire Limestone > Chalk > Permo-Triassic sandstone. Mean groundwater concentrations however exhibit a slightly different trend with Permo-Triassic sandstone > Lower Greensand > Lincolnshire Limestone > Chalk. In the case of the Permo-Triassic sandstone the relatively high mean value is the result of one very high concentration taken from a confined part of the aquifer and so may not be representative of methane concentrations in this aquifer as a whole. For this reason, subsequent calculations use the median concentration rather than the mean.

Figure 2 shows cumulative frequency plots for the four main aquifers and demonstrates that the data for three of the aquifers is normally or log normally distributed. Data from the Permo-Triassic sandstone suggest that there are possibly two or three populations: one below the detection limit, a second between 1 and 3 µg/l and possibly a third population with concentrations greater than 10 µg/l. Although there is a paucity of data for the Permo-Triassic aquifer, Figure 2 acts as an indicator of possible trends within the data-set.

Crystalline rocks

Table 3 shows CH_4 concentration data for four sites in Scotland: two in Caledonian granodiorites and two in metasediments of Precambrian and Dalradian age. Methane at three of the sites was below detection, and only just above at the remaining site. These values are little different from the equilibrium concentration of CH_4 expected for water in contact with the atmosphere.

High-methane waters

As mentioned earlier, moderately high CH_4 concentrations are found where groundwaters are most reducing, normally in confined conditions. Such conditions are often accompanied by sluggish or even no discernible water circulation: an example of the former is found in the down-dip portion of the Lincolnshire Limestone, while the latter occurs in the concealed Triassic sandstone strata of the Wessex Basin (Table 4). Strongly-reducing waters tend not to be exploited except for very specific purposes such as geothermal heating, and natural discharges are likely to be minimal.

The highest CH_4 concentrations, however, appear to be associated with the tunnelling of mudrocks. Figure 3 shows results obtained from inflows within two transfer tunnels, Carsington (data from Pearson & Edwards, 1991), and Wyresdale (data from Bath et al., 1988), both excavated in Namurian strata. Amounts of methane in solution reach peaks of 15,000 and 13,000 µg/l in the respective tunnels, equivalent to some 20 and 180 cc STP per litre of water. These concentrations equate to CH_4 partial pressures of approximately 0.5 and 4 bars respectively.

DISCUSSION

Methane Formation

Biogenic methane formation is the most likely source in these aquifers and so it is reasonable to expect the CH_4 concentration to be in part explained by the availability of organic carbon undergoing methanogenesis. However, there is a lack of published data for the organic carbon content of UK aquifers. Harrold et al. (2003) provide some values for Chalk and Permo-Triassic sandstones reporting organic carbon concentrations as between 0.1- 0.2% for the Chalk and <0.05 - 0.07% for the sandstones. This probably explains the generally low methane values observed in these aquifers and may even provide an explanation as to the observed higher median methane concentrations in the Chalk compared with the Permo-Triassic sandstone (Table 2). The origin of methane in English groundwaters is the subject of a further study (Darling & Gooddy, in preparation).

Calculation of methane emissions

Indirect emissions of nitrous oxide from regional aquifers have previously been determined by Hiscock et al. (2003). This study has used a similar approach by obtaining estimates for annual abstraction from each of the aquifers (m^3/yr) (Downing, 1993), converting this to l/yr ($m^3 \times 10^3$), multiplying this value by the mean, median or maximum methane concentration for each aquifer type (μg/l) and finally converting this to Tg/yr (μg $\times 10^{-18}$). The method assumes that the methane will completely outgas once it has been abstracted which is likely to lead to an over-estimation of methane emissions.

Table 5 shows the total emissions of methane based on abstraction volumes from each of the four aquifer types. On this basis the Chalk groundwaters are the greatest source of methane owing to the much higher abstraction volumes. For each of the aquifers there will be an element of natural discharge, which might perhaps double the abstraction emissions.

Groundwater in crystalline rocks appears to contribute nothing of significance to CH_4 emissions, which is fortunate since any figure for discharge would be highly speculative. However, high-CH_4 groundwater discharges, though small in volume are orders of magnitude higher in concentration than potable water sources (Figure 2). These outputs are not well characterised but it is conceivable that emissions are of the same order as those from the major aquifers.

By summing the calculated methane emissions from groundwater abstraction volumes and the maximum measured methane concentrations; an equal amount of emissions resulting from natural discharge; and an equal amount from high CH_4 groundwater discharges that are very low in volume, the total CH_4 emission from UK groundwaters might therefore total 1.0 x 10^{-3} Tg CH_4/yr (i.e. 3 times as much as the calculated maximum abstraction emission). Based on the values of Salway et al. (2002) who estimated a total UK emission of 2.2 Tg CH_4/yr, the groundwater contribution accounts for no more than 0.05% of this. To put this contribution into context, UK CH_4 emissions are mostly accounted for by agricultural activities (37%), followed by landfill (22%), gas leaks (16%) and coal mines (11%) (Salway et al., 1994). In terms of a global methane budget of 587 Tg CH_4/yr (Hein et al., 1997) the methane contribution from UK groundwaters is a further two orders of magnitude lower.

Methane Hazards

It is clear from the data summarised above that most groundwaters in the UK, potable or otherwise, have CH_4 contents orders of magnitude below the theoretical 1600 μg/l that is necessary to reach the lower explosive limit after outgassing in a confined space. In reality it is likely that significantly higher CH_4 concentrations are required before an explosion hazard results. Such waters do exist in aquifers where conditions are suitably reducing: the examples from the down-gradient Lincolnshire Limestone aquifer and the concealed Sherwood Sandstone are cases in point. In particular, however, it is the waters from organic-rich formations generally regarded as aquitards that pose the greatest risk when pressure is released by excavation, in particular tunnelling. Fortunately this risk is now well recognised so that the appropriate measures can be taken (Hooker and Bannon, 1993).

CONCLUSIONS

Samples of groundwater from a range of rock types across the UK have been analysed for their methane content. Even using ‘worst case’ estimates of groundwater-derived emissions, it is clear from the data that such sources cannot be a significant contributor to the UK, let alone global, methane budget.

Nevertheless, against the generally low background concentrations typical of the UK’s main aquifers certain instances of very high methane waters stand out. These hazardous emissions are usually associated with Upper Carboniferous strata, but it is highly probable that tunnelling in organic-rich clays of any age would produce a similar outcome.

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Table 1. Infiltration and abstraction of major UK aquifers in million m^3 per year (after Downing, 1993)

	Infiltration	**Abstraction in 1977**
Chalk	4631	1255
Lower Greensand	275	86
Lincolnshire Limestone	86	43
Triassic sandstone	1443	587

Table 2. Statistical summary of methane data from potable groundwaters

	Chalk	**Lower Greensand**	**Lincolnshire Limestone**	**Triassic Sandstone**
Samples	44	20	9	12
Mean (µg/l)	2.81	7.89	4.42	39.5
Median (µg/l)	0.94	7.66	2.60	0.465
Standard deviation (µg/l)	7.17	6.27	6.50	134
Minimum (µg/l)	<0.05	<0.05	0.05	<0.05
Maximum (µg/l)	42.9	22.0	21.2	465
Confidence level (95.0%)	2.18	2.94	5.00	85.2

Table 3. Methane concentrations from crystalline rocks in Scotland

Site	Rock Type	Methane (µg/l)
Cruachan	Caledonian granodiorite	<0.05
Foyers	Caledonian granodiorite	0.09
Glenmoriston	Moine metasediments	<0.05
South Lawers	Dalradian metasediments	<0.05

Table 4. High methane concentration groundwaters not from public-supply sources

Site	**Rock Type**	**Methane** (μg/l)
Stow 4[a]	Lincolnshire Limestone	1700
Stow 6[a]	Lincolnshire Limestone	2300
Marchwood geothermal	Sherwood Sandstone, Southampton	7790
Western Esplanade geothermal	Sherwood Sandstone, Southampton	5540

[a] Data from Bishop and Lloyd, 1990

Table 5. Atmospheric emissions of methane based on abstractions from potable UK groundwaters

	Abstraction (m^3/yr)	**Methane** (µg/l)		**Emissions** (Tg/yr)	
		Median	Max	Median	Max
Chalk	1.26×10^9	0.94	42.9	1.17×10^{-6}	5.38×10^{-6}
Lower Greensand	8.60×10^7	7.66	22.0	6.59×10^{-7}	1.89×10^{-6}
Lincolnshire Limestone	4.30×10^7	2.60	21.2	1.12×10^{-7}	9.12×10^{-7}
Triassic sandstone	5.87×10^8	0.47	465	2.73×10^{-7}	2.73×10^{-4}
SUM	1.97×10^9			2.22×10^{-6}	3.30×10^{-4}