

Future Fugitives

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Introduction

Coal is the mainstay of electricity production throughout almost the entire world – a situation likely to be the case into the foreseeable future, though its proportion may go down as gas and nuclear energy increase. Whether wind and solar become a significant player is very uncertain at this stage. Coal (as coking coal) is also used in steel production. Apart from generating CO₂ when it is oxidised in the power station or in steel making, another powerful greenhouse gas, methane, is inadvertently released whilst mining the coal. This gas, called coal seam gas (or coalbed methane, CBM, though this term is more often restricted to unmined coal) is adsorbed within the coal mass and released as the overburden pressure is removed by mining and the coal broken up during excavation. Such emissions are called fugitive emissions and can be a significant fraction of a mining country's methane account.

First, I will present an overview of gas in coal in terms of its origin and how seam gas content varies with depth. Then I discuss factors that influence the degree of fugitive emissions released by mining and how these are factored into the inventory procedure before presenting some estimates of the global release of methane, broken down by major mining countries. Finally I will present some estimates of fugitive emissions from Indian coal mining to 2030 based on the predicted coal demand.

Gas in coal

Composition

Coal seam gas consists usually of methane ~90+% (sometimes with small amounts of higher hydrocarbons such as ethane and propane) plus some nitrogen and carbon dioxide (CO₂). In some coalmines, particularly in Australia, but also in France and Poland, elevated levels of CO₂ are encountered which can approach 100%. CO₂ poses a different danger to CH₄ in underground mining, with outbursting tending to occur at lower seam gas contents and is rapidly toxic at high concentrations. Occasionally, toxic levels of H₂S can be present.

Origins

Methane

The standard view of how methane got into coal is that it is generated in the coalification process. Decaying vegetation laid down in swampy anoxic conditions initially forms peat by loss of CO₂ and CH₄ as in a landfill, then if the peat sediment is more deeply buried it is transformed to lignite through further loss of oxygen then through low rank coal to high rank coal and anthracite. These transformations occur on geologic timescales and their extent is a function of depth of burial which increase the rate of the coalification process due to increased temperature. The progress of coalification can be depicted, schematically, for humic coals in Figure 1 which shows how the elemental composition of the coalifying material changes with time in the form of a van Krevelen diagram. In the early stages the oxygen content drops due to loss of CO₂ and H₂O (with the loss of H and C roughly balancing each other out), then loss of hydrogen becomes apparent with the onset of methane emissions. The overall process can be described by an Arrhenius-style rate equation and it can be simulated in the laboratory by encapsulating peat and heating to ~150⁰C or more for a few years. Note that there are some waxy plant precursors that start off with much higher H/C and

lower O/C ratios to form sapropelic coal which can be found alongside humic coal. Cannel coal is an example.

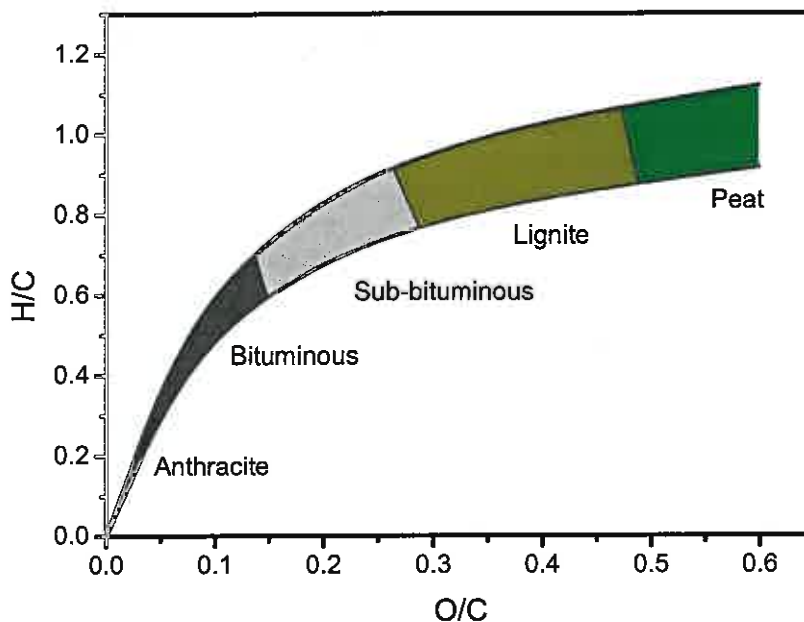


Figure 1. Changes in elemental ratios during the coalification process

It seems that this picture is not the full story. Studies of the C^{13} isotopic composition of coal seam methane are often not consistent with what would be expected from the thermogenic conversion of vegetable matter to coal (see for example Faiz and Hendry, 2006). The levels of C^{13} can vary slightly in carbon-containing material due to differences in the reaction rates involving C^{12} and C^{13} , the lighter isotope tending to react faster. This is particularly marked for microbial or enzymatic processes. The amount of C present as C^{13} is usually expressed as a deviation, δ , expressed in parts per thousand (‰), from a standard material (Pee Dee Belemnite or PDB – a carbonate mineral). Even at low temperatures characteristic of deep burial, thermogenesis would produce CH_4 with a PDB δC^{13} content in the range -35 ‰ to -20 ‰ (Cramer et al., 2001) but a number of studies have found δ values of -50 ‰ and less. This is characteristic of microbially generated CH_4 from landfill. Microbes have been found at considerable depths in the earth's surface and this has led to the suggestion that coal, being porous and often full of water, allows fermentation of organic acids present within the coaly aquifer to produce CH_4 . The assumption is that all or part of the original thermogenic methane has been displaced by this microbial gas.

In the absence of oxygen, microbes can reduce either CO_2 to produce CH_4 and water or split acetate (acetoclastis) to CH_4 and CO_2 . The deuterium content in CH_4 can shed further light on to the origins of coalbed methane which provides another dimension to the C^{13} data as is shown in Figure 2.

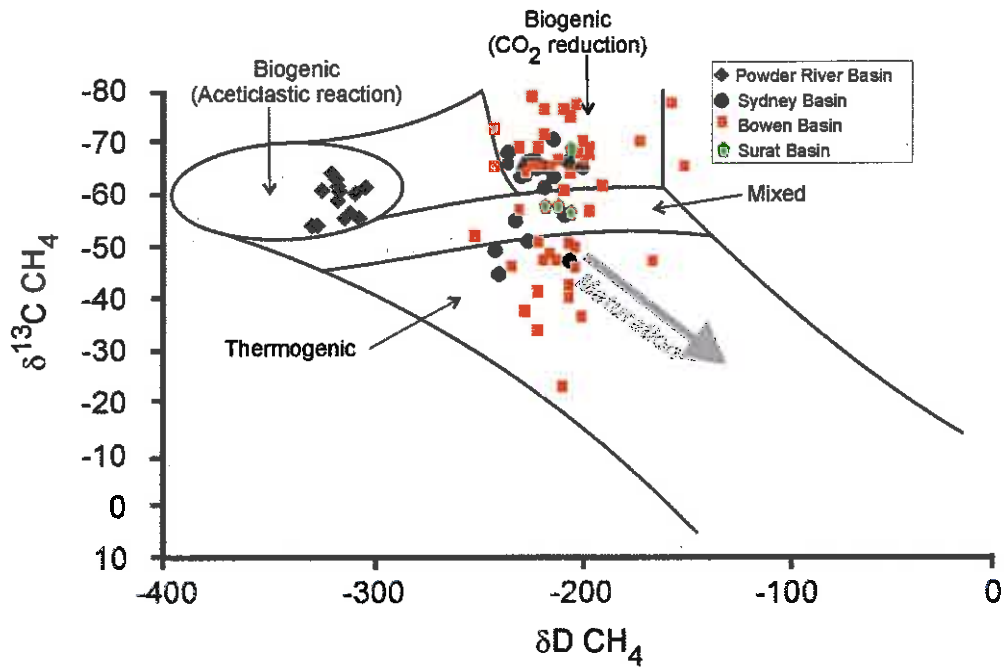


Figure 2. Data on the distribution of stable isotopes in coal seam methane
Source Faiz and Hendry (2006)

In deeper coal seams, CH₄ appears to be more thermogenic in origin (Figure 3)

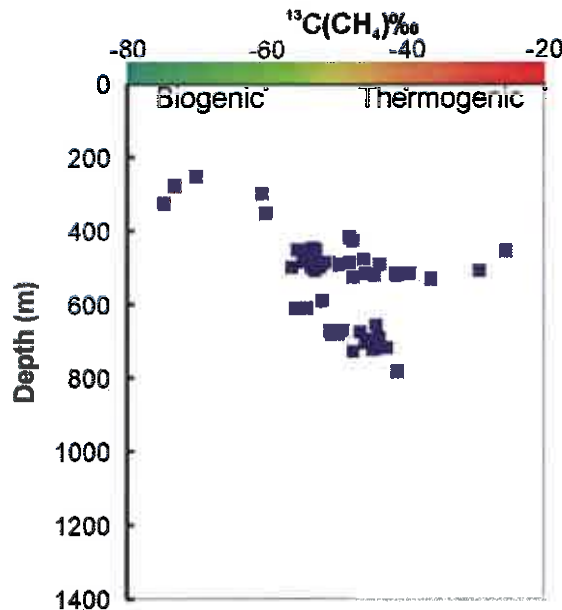


Figure 3. Variation in C¹³ with seam depth for an Australian coal district.
Source Faiz and Hendry (2006)

Recently, some Chinese workers (Qin Shengfei et al., 2006) suggested that methane light in C¹³ could result from adsorption/desorption processes within the wet coal with the heavier isotope being slightly more soluble due to its greater polarity. They observed that, in seams with more active hydrology, the gas was lighter than that from more stagnant water flow. They also noted that this was consistent with their finding that dissolved CH₄ found in water associated with gas or oil basins was heavier than the source gas. It should be borne in mind however that deuterated methane is less soluble which could mitigate the effect somewhat (Bacsiz et al., 2002). Nevertheless, this is an interesting theory.

Carbon dioxide.

CO₂ seam gas is believed to originate from upwelling magma permeating through faults (Smith et al., 1985). Indeed, high CO₂ seam gas can be found on one side of a fault whilst CH₄ predominates on the other. Usually the high CO₂ contents occur in the upper coal seams.

Gas content and depth.

It is well-known to underground miners that the deeper one mines, the gassier the coal seems to get. Indeed, eventually one has to pre-drain the coal seam to minimise the risk of outbursts and explosions due to the high gas pressure difference across the exposed face of the coal. In Australia pre-drainage has to be performed when CH₄ contents exceed 9 m³/tonne (6 m³/tonne for CO₂). The affinity of coal for CH₄ increases with pressure and can be measured in the laboratory as an adsorption isotherm. The coal is powdered and the degree of uptake can be measured either gravimetrically or volumetrically at various pressures. An example is shown in Figure 4. Note the data are shown in terms of kg/tonne CH₄ whereas the more usual units are m³/tonne (1 kg/tonne ~1.5 m³/tonne).

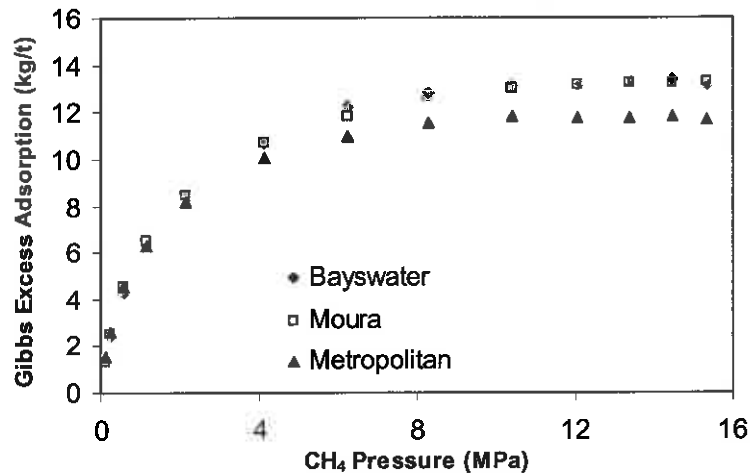


Figure 4. CH₄ adsorption isotherms for 3 Australian coals

Is this what we get in practice when measuring gas contents as a function of depth? Not really as the data of Faiz et al (2007) illustrate in Figure 5. The red points provide an indication of typical methane adsorption capacity based on isotherm measurements similar to those in Figure 4.

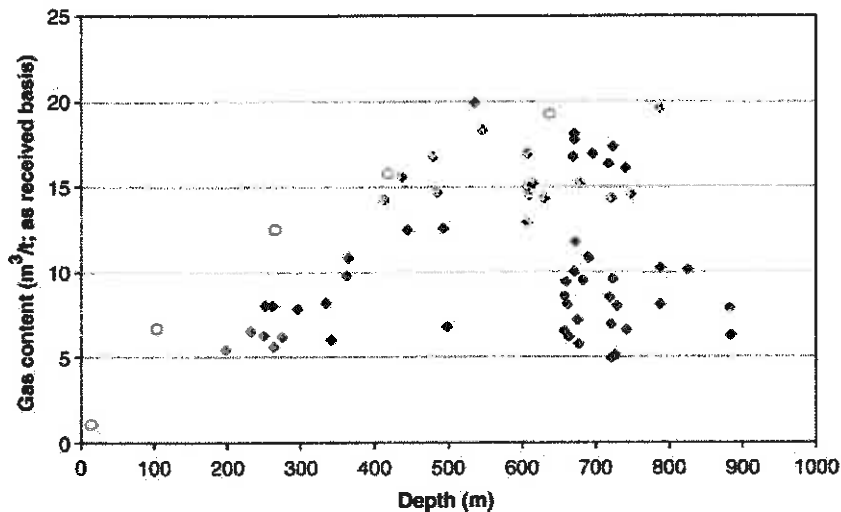


Figure 5. In-situ gas contents as a function of depth for some Sydney Basin coals

At shallow depths one would expect gas contents to be lower than isotherm data due to gradual loss to the surface. However, the decrease in gas content at depths of ~700m or more is, perhaps, unexpected. However, the isotherm data are for one particular coal whereas the gas content data come from coal of different depths and hence quality – rank usually increases with depth. Adsorption capacity first increases as rank increases then drops off. The deeper coals are higher in vitrinite which, having less surface area, has less adsorption capacity. The ash contents of coals in Figure 5 were generally in the range 5-10% with three samples greater than 20%. The ash effectively dilutes the coal and its gas content potential. If one plots the maximum gas contents that have been found as a function of depth for coal worldwide (listed in Sloss, 2005), they would lie along the upper envelope of the data in Figure 5 but drop less slowly after 800m being still ~ 15 m³/t at 1000m depth at ~ 10 m³/t at 2000m. However, minimum gas content values at all depths are close to zero except at 1900m or more where they are about 3 m³/t, but there are very few measurements. One can conclude, therefore, that depth is not that good a guide to gas content.

Extent of fugitive emissions

Let me turn now to fugitive emissions. The reason for their importance is that most of such emissions consist of CH₄ which is a potent greenhouse gas. The threat of climate change is forcing the world's nations to count their contribution to the problem. This is being done through the Intergovernmental Panel on Climate Change (IPCC) as part of the United Nations Framework Convention on Climate Change (UNFCCC). The guidelines for preparing inventories have just been updated from the 1996 version.

The procedure is classified according to detail:

- Tier 1 – calculates emissions based on a national activity with a single national emission factor (EF) which maybe one recommended in the guidelines (ie a global EF).
- Tier 2 – calculates emissions from the sum of regional activities each with a regional EF. Note that the regions boundaries may not be administrative or political.
- Tier 3 – calculates emissions at the individual enterprise level with an enterprise-specific EF.

The extent of fugitive emissions from coal mining is greater than that expected from just the gas contained in the coal seam being extracted. This is because mining effects neighbouring seams as indicated in Figure 6. Gas is released from the target seam, another seam below becomes fractured or relaxed and from exposed faces in the high wall. Also some carbonaceous material is dumped into the spoil which can continue to emit CH₄ for awhile.

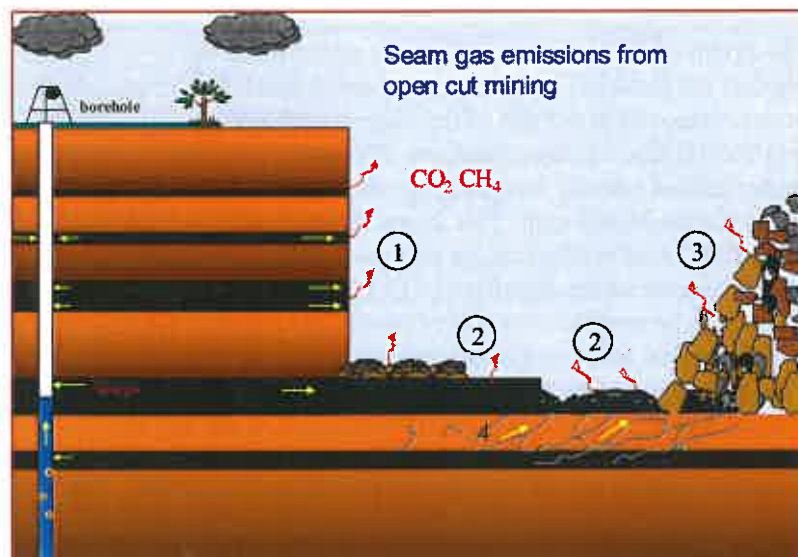


Figure 6. Release of fugitive emissions from an opencut

Measuring or estimating fugitive emissions is a challenging task. Only recently have mining companies started to routinely measure gas contents of coal seams from exploration boreholes on opencut mines which can allow the size of the gas reservoir to be estimated. The borehole in Figure 6 was part of a study designed to quantify such emissions (Saghafi et al., 2003). Prior to then I and my colleagues have carried out measurements of the cross-wind concentrations of CH₄ within the plume leaving an opencut mine using mobile air pollution instrumentation (Williams et al., 1993). These measurements demonstrated that more CH₄ was emitted per tonne of coal mined in NSW opencuts than in Queensland and they form the basis of the Australian methodology for opencut fugitive emissions.

An opencut mine can, in principle, extract all coal sequences above the limiting depth of opencut mining (currently ~180-200m), so the extra fugitive release could be relatively small. An underground mine, by contrast, only excavates one seam and will disturb seams above and below which can't be subsequently mined as is shown schematically in Figure 7 in which the red seams denote other coal or carbonaceous strata releasing their seam gas into the level of the mine workings.

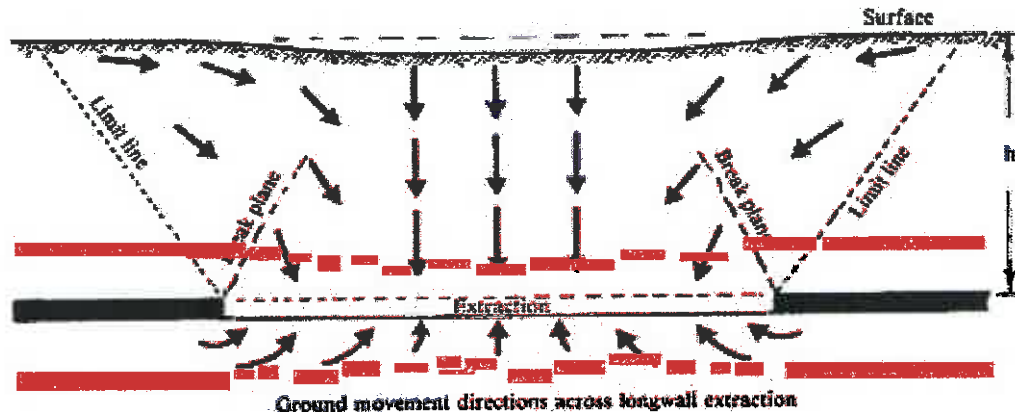


Figure 7. Schematic across an excavated longwall panel showing the ground disturbance above and below the mined seam.

Consequently, the extent of fugitive emissions to be added on to the in-situ gas of the mined coal is high. Note that for bord and pillar mining, there is much less ground disturbance (unless the pillars are removed at the end of the mine's life) and consequently the extra fugitive emissions will be less. Measurements by Williams et al, (1992, 1996) found that, on average, gassy underground (mostly longwall) mines emitted more than four times the amount of in-situ gas in the mined coal. This forms the basis of the Australian methodology for gassy mines, whereby coal production, in-situ gas content and an emissions multiplier constitute the emissions estimation algorithm (AGO 2006).

Methane continues to be emitted even after the coal has reached the surface. Although emission factors are variable for post-mining underground coal (depending on gas content at time of mining, elapsed time before reaching the surface, lump size etc), it can average out to 1-2 m³/tonne (Williams et al., 1993) and should be included in the inventory.

In principle, as the gas from underground mines comes out of a pipe, albeit a big one and as, in Australia at least, such mines must measure the CH₄ content then a Tier 3 approach is quite feasible using ventilation air flows and CH₄ concentration data with due allowance for any pre-drainage and utilisation.

The global picture

Figure 8a illustrates the growth of fugitive emissions from the top 10 coal producers in 1990 (USEPA, 1994). From this one might deduce that the apparent non-linear increase in emissions as tonnage increases is due to an increasing contribution from more gassy underground mines. However this may be in error as plotting the overall release of CH_4 from just the underground production gives a linear plot.

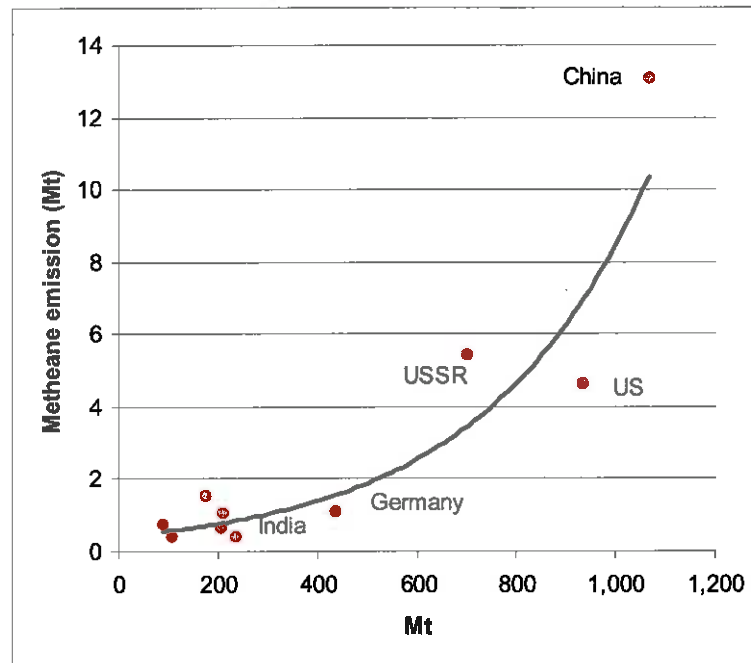


Figure 8a. Fugitive methane emissions from the world's top ten coal producing countries as a function of overall production for 1990.

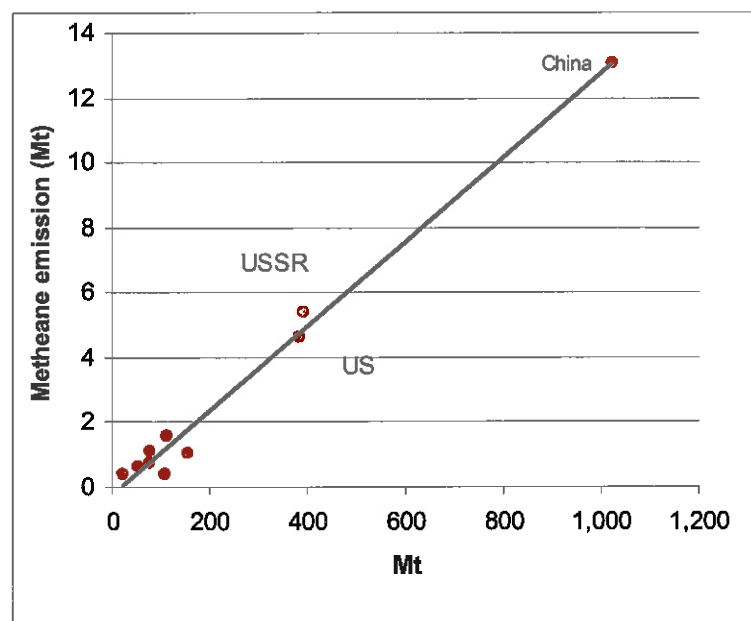


Figure 8b. Fugitive methane emissions from the world's top ten coal producing countries as a function of underground production for 1990.

The bulk of China and USSR production came from underground.

If we take emission data for 2004, we get a somewhat different picture (Figure 9) which seems to indicate that emissions increase pro rata with production. I have no information concerning the split between underground and opencut production

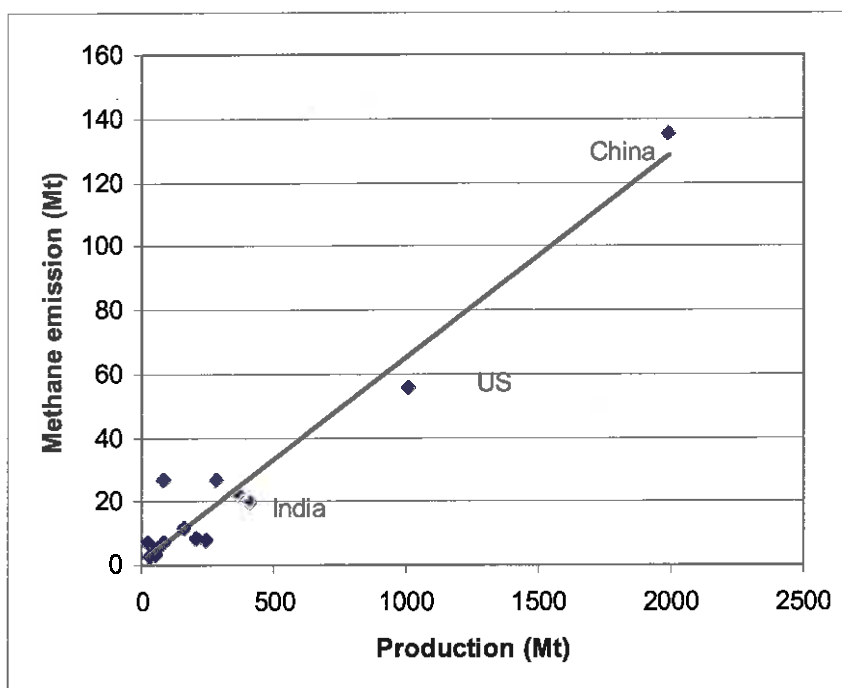


Figure 9. Fugitive methane emissions from the world's top coal producing countries as a function of production for 2004.

(source http://iea.org/textbase/papers/2007/methane_fact.pdf; BP 2007)

Overall the data may reflect the variability in gassiness and the uncertainty in emission quantification.

The Indian situation.

Many developing countries have difficulty in preparing a detailed greenhouse gas inventory for a number of reasons. These include lack of sufficient recorded data on a given activity and poor knowledge of local emission factors, private companies unwilling to provide data which might compromise confidentiality or competitive advantage, the size of the informal economy etc. India is no different in this regard, indeed these constraints are outlined in India's first (and the only) National Communication to the UNFCCC covering the base year 1994 (GoI 2004). Thus we should look at the Indian estimates with this in mind.

For 1994, it was estimated that 650 Gg (the reporting units for GHG inventories, note 1Gg = 1000 tonnes) of CH₄ came from fugitive emissions; that is coal fugitives represented about 3.4% of total CH₄ emissions, most of which came from cattle and rice cultivation. Coal mining amounted to 253 million tonnes (Mt) which translates to an overall EF of 3.8 m³/t. This can be contrasted to data for Australia for the year 2005 whereby 1029 Gg CH₄ came from 399 Mt raw coal representing an EF of 3.9 m³/t (or 5.2% on a saleable coal basis). These EFs are a composite of both underground and opencut emissions. For India, opencut mining accounted for 75% of total production whilst for Australia it was very similar at 78%. In the Australian inventory, opencut EFs range from 1.2 m³/t for Queensland to 3.2 m³/t for NSW, averaging at about 2 m³/t. Thus the EF for underground production is ~ 10 m³/t on a raw coal basis and a similar figure would apply for 1994 Indian underground coal if the same opencut EF is used. However, much of India's underground coal comes from hundreds of small bord and pillar operations which are unlikely to be very deep, whereas there are only about 40 underground mines in Australia, mostly longwall, some at depths of 400m and gassy. Also

assuming the Indian production figures are for raw coal which has high mineral matter content, I would expect an average underground EF to be lower, perhaps no more than ~5 m³/t.

Another estimate for CH₄ emissions from Indian coal mining for the year 1994 can be found in a World Bank publication (<http://www.worldbank.org/html/pic/indiacol.htm>) amounting to 1800Gg which would give an overall EF (ie including opencut) of about 10m³/t. From the above discussion, I think this is quite unrealistic. Obviously – at least on the basis of the information available to me - more studies need to be done.

Future projections.

The path to the developed world seems to be one of increasing energy use and availability. Most of this is sourced from primary fossil fuels with coal the mainstay for stationary energy and oil for transport although natural gas is making inroads in both these sectors. Until now, India's demand for energy has been increasing exponentially, a trend likely to continue. There have been a number of projections for energy demand which vary significantly according to the in-built assumptions particularly growth rates. Here are two of them, note that the data are expressed in terms of Mt oil equivalent (Mtoe) the amount of oil needed to provide the same amount of primary energy – for coal one has to multiply by 1.5 to get tonnages of coal:

Table 1a. Predicted primary energy demand by source for India (Mtoe)

	Year			
	1990	2004	2015	2030
Coal	105	196	283 (259)	450 (338)
Oil	63	127	184 (176)	268 (238)
Gas	10	23	40 (39)	68 (63)
Nuclear	2	4	18 (19)	36 (48)
Hydro	6	7	12 (13)	21 (25)
Biomass/waste	176	214	236 (224)	253 (241)
Other	0	0	2 (3)	7 (10)
Total	361	573	776 (733)	1104 (964)

Source IEA (2006)

Table 1b. Predicted primary energy demand by source for India (Mtoe)

	Year			
	2002	2012	2022	2032
Coal	228	289	393	970
Oil + Gas	150	232	325	475
Nuclear	5	16	52.5	110
Hydro	20	70	115	150
Other renewables	0	10	40	50
Total	403	617	925.5	1755

Source: Grover and Chandra (2006)

The first of these tables includes allowances for increased energy efficiencies, the values in parentheses assume greater efficiency increases forced by shortage of oil whilst the second is more in line with India Vision 2020 estimates. It can be seen that there is almost a factor of 2 difference by 2030. If we couple this with the statement by the Indian Minister for Coal that India could require over 2000 Mt coal (1300 Mtoe) by 2031 (Reuters 2007) then we can see the crystal ball is a little cloudy.

Using these estimates to predict fugitive emissions makes the ball even cloudier:

- will the demand be largely satisfied from within?
- if so will the split between opencut and underground remain similar?
- will underground mining transform to highly mechanised longwall operations?

However, from Figures 8a and 9, we can see that, by and large for the big miners, fugitive emissions go up in proportion to coal production. If we use the EF generated for the 1994 Indian inventory, then we get the following estimates for 2015 and 2030 for the two scenarios. For the Grover and Chandra (G&C) case, the levels of fugitive emissions begin to approach that currently emitted by rice cultivation, the second largest emitter of CH₄.

Table 2. Estimated fugitive emissions from black coal mining in India

	2015	2030
IEA	1075	1710
G & C	1140	3610

Concluding remarks.

The growth in the Indian coal demand will inevitably bring with it a concomitant increase in fugitive CH₄ emissions. The degree will depend on the mining technology deployed, particularly for underground mines, but it will be a significant quantity from a climate change perspective. Currently, knowledge of the extent of such emissions for India is shaky; indeed the very word 'fugitive' implies uncertainty. However, emissions from underground mines can be measured relatively easily and with reasonable accuracy but those from opencut operations remain a challenge. We are likely to see a price put on greenhouse gas emissions which could well impact on mine economics in a serious way. This will drive studies designed to reduce the uncertainty of fugitive emissions inventory and projects to utilise these emissions. At this stage only emissions from underground mining are amenable to mitigation. Pre-drained gas ahead of mining can be a) flared thereby reducing the impact by a factor of ~7, b) fed into a natural gas pipeline after conditioning or c) utilised to generate electricity in either reciprocating engines or turbines – these technologies are well established. Reducing CH₄ in ventilation return is much more problematic; perhaps the only feasible process currently is catalytic oxidation in a device put immediately after the exit of the ventilation system. Provided CH₄ levels exceed 0.1-0.2% this can be self-sustaining energy-wise and at higher CH₄ concentrations can become a net power source. One such device (a Megtec VOCsidizer®) is being put in place in an Australian mine designed to handle 20% of the ventilation return flow.

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References

- AGO 2006. Australian Methodology for the Estimation of Greenhouse Gas Emissions and Sinks 2004. Australian Greenhouse Office.
- Bacsik Z., Canongia Lopes J.N., Costa Gomes M.F., Jancsó G., Mink J. and H. Pádua A.A. 2002. Solubility Isotope Effects in Aqueous Solutions of Methane. *Journal of Chemical Physics*, 116, No 24, 10816-10824
- BP 2007. Statistical Review of World Energy 2007. Available from <http://www.bp.com/productlanding.do?categoryId=6848&contentId=7033471>
- Faiz M and Hendry P (2006) Significance of Microbial Activity in Australian Coal Bed Methane Reservoirs - a Review. *Bulletin of Canadian Petroleum Geology*, 54, No 3, 261-272
- Faiz M., Saghafi A, Sherwood N and Wang I. 2007. The Influence of Petrological Properties and Burial History on Coal Seam Methane Reservoir Characterisation, Sydney Basin, Australia. *International Journal of Coal Geology* 70, 193–208
- GoI. 2004. India's Initial National Communication to the United Nations Framework Convention on Climate Change. Ministry of Environment and Forest, Government of India. pp292.

- Grover, R.B. and Chandra, S., 2006. Scenario for Growth of Electricity in India. *Energy Policy*, 34, 2834-2847
- IEA, 2006. World Energy Outlook 2006. International Energy Agency, Paris, pp 600
- Qin Shengfei, Tang Xiuyi, Song Yan and Wang Hongyan. 2006. Distribution and Fractionation Mechanism of Stable Carbon Isotope of Coalbed Methane. *Science in China Series D: Earth Sciences*, 49, No12, 1252-1258
- Reuters, 2007. 'India's Coal Demand May Quadruple by 2031 -- Minister'. Available at www.planetark.org/dailynewsstory.cfm/newsid/40808/story.htm -accessed May 2007
- Sloss L. 2005. Coalbed Methane Emissions –Capture and Utilisation. IEA Clean Coal Centre 56p.
- Smtih J.W., Gould K.W., Hart G. and Rigby D. 1985. Isotopic studies of Australian Natural and coal seam gases. *Proc. Australian Institute of Mining and Metallurgy*. 290, 42-51.
- Williams D.J., Carras J.N., Saghafi A., Lange A., Thomson C.J., Francey R.J., Steele L.P., Langenfelds R.L. & Fraser P.J. 1992, Measurement of the Fluxes and Isotopic Composition of Methane Emissions., Energy Research and Development Corporation, End-of-grant Report, Project 1460.
- Williams D.J., Saghafi A., Lange A., & Drummond, M.S. 1993, Methane Emissions from Open-Cut Mines and Post-Mining Emissions from Underground Coal, CSIRO Investigation Report, CET/IR173.
- Williams D.J., Saghafi A., & Lama, R.D.. 1996, Methane Emissions From Coal Mining. International Energy Agency, IEA Greenhouse R & D Report PH2/5, Paris.