ATMOSPHERE

## Methane on the Rise-Again

Euan G. Nisbet<sup>1</sup>, Edward J. Dlugokencky<sup>2</sup>, Philippe Bousquet<sup>3</sup>

Methane is the most unpredictable of the greenhouse gases. Roughly a fifth of the increase in radiative forcing by human-linked greenhouse gases since 1750 is due to methane. The past three decades have seen prolonged periods of increasing atmospheric methane, but the growth rate was slowing (1) and from 1999-2006 the total amount of methane in the air (the 'burden') was nearly constant. But from 2007, strong growth has returned. The reasons for these observed changes remain poorly understood because of our limited knowledge of what controls the global methane budget (2).

Estimates of methane emissions vary widely; global estimates derived from process studies of sources (termed 'bottom-up') are generally much larger than from direct observation of the air (termed 'top-down') (2). Methane sources are about 2/3 anthropogenic and 1/3 natural. Many local industrial emissions may be significantly underestimated (3). The renewed rise in the methane burden prompts urgent questions about the underlying causes, but globally, *in situ* monitoring to track atmospheric methane is very limited outside the major nations.

Methane sources and sinks vary with latitude. At polar latitudes, methane sources include wetlands, some of the world's most important natural gas wells and pipelines, thawing permafrost, and methane hydrates, an ice-like substance that can store huge amounts of methane. In the heavily populated northern mid-latitudes, the main sources are the gas and coal industries, agriculture and landfills, and biomass fires. Tropical wetlands are the world's largest natural source of methane (4). Emissions from equatorial and savanna wetlands, ruminants, and biomass burning are increased further by tropical anthropogenic inputs, for example in southeast Asia. The main methane sink is reaction with OH, especially in the tropical mid-troposphere. Minor sinks include soil oxidation, reaction with marine chlorine, and reactions in the stratosphere.

From the 1980s until about 1992, atmospheric methane was rising sharply by about 12 parts per billion (ppb) per year (see 'Methane ups and downs', panel A). Then came a decade of much slower growth, about 3 ppb/year, coupled with a sudden decrease in the north-south interpolar difference (1). In the early 2000s, growth almost ceased and there were short periods when the burden declined, in 2000, 2001 and 2004. In the 'stagnation' period 1999-2006, these top-down findings from atmospheric data differ markedly (5) from bottom-up inventories, which detail strong growth in anthropogenic emissions. Yet, although not well constrained, the main sink— oxidation by OH radicals—seems little changed (4). This discrepancy between 'top-down' and 'bottom-up' budgets remains unresolved.

In 2007, just when scientists thought methane had stabilized, it rose again and since then global average growth has been strong, at about 6 ppb/year. Considering the latitudinal zones in more detail (see 'Methane growth rate by latitude' Panel B), Arctic methane rose dramatically in 2007 but since then Arctic growth has tracked global trends. Large emissions attributed to decaying methane hydrates in permafrost have been reported from waters of the East Siberian Arctic Shelf (6), but are not apparent in US NOAA atmospheric observations (see Panel B) nor detected in isotopic measurements from surface and aircraft sampling in the European Arctic (7), which point to wetland as a major Arctic source in summer and industrial gas leaks in winter. Long-term release of methane from hydrate is probable (8), but catastrophic hydrate emission scenarios (9) are unlikely.

In the southern tropics, sustained growth above global trends has occurred from 2007 (Panel B). For example, at Ascension Island (8°S) which samples the tropical South Atlantic, growth was about 10 ppb per year from 2010 to 2011 during a period of wet regional summers when wetlands will have expanded. This rise in natural emissions of methane, regionally sustained over five years, is particularly interesting because it may also give insight into past glacial terminations and initiations, when the methane burden changed sharply, perhaps from similar processes.

Atmospheric data show that global emissions have risen by about 15 to 20 Tg (million tons) per year in the past half-decade. Global-scale modeling of these methane observations (4, 5, 10) suggests that in 2007, tropical wetland emissions dominated the increase, although high northern latitude output was also important. Since then, most of the increase has been driven by the tropics (9 to 14 Tg/year) and northern mid-latitudes (6 to 8 Tg/year) (10).

Since 2007, there is much to suggest that emissions from human activities have also increased. In particular, world natural gas leaks may have increased as consumption has grown (11), for example in the US, China and Japan (which is sharply increasing gas imports to replace nuclear power). In the US, which has overtaken Russia as the largest gas producer (11), hydraulic fracturing is increasingly important. In Utah, fracking may locally leak as much as 6 to 12% of gas production to the air (12) but a full understanding of fracking's greenhouse impact demands monitoring over the full gas-well lifetime and analysis of the transport distribution system. Global coal mining has also dramatically expanded (11), especially in China, where many mines are notoriously gassy. Rising energy production suggests increased emission, but this inference needs to be reconciled with observational data on <sup>13</sup>C in methane. Since 2007 atmospheric CH<sub>4</sub> has become more depleted in <sup>13</sup>C, an indication that growth is dominated by wetland and ruminant emissions, which are rich in <sup>12</sup>C.

More data are needed to resolve top-down vs. bottom-up divergence but the measurement network for methane concentration and isotopes is very thin. During a debate on the methane problem at a meeting of the European Pergamon Arctic methane group in Kiel, Germany in November 2013, Patrick Crill commented that "data without models are chaos, but models without data are fantasy." Spatially and temporally, better measurement is essential to identify and quantify methane sources (3, 4, 10), but long-term data gathering is in trouble. Despite methane's attractiveness as a cost-effective greenhouse reduction target, US budgets for greenhouse gas monitoring are contracting, while in situ methane measurement barely shows over the terminator of Europe's new €80 billion "Horizon 2020", with low priority. Somewhere, perhaps in the tropics or East Asia, unwelcome methane surprises may lurk, but watchers are few.

## **References and notes**

1. E. J. Dlugokencky et al., Phil. Trans, R. Soc. Lond. A 369, 2058 (2011). 2. S. Kirschke, Nat. Geosci. 6, 813 (2013). 3. S. M. Miller et al., Proc. Natl. Acad. Sci. U.S.A. doi10.1073/pnas.1314392110. 4. P. Bousquet et al., Atmos. Chem. Phys. 11, 3689 (2011). 5. I Pison et al., Atmos. Chem. Phys. 13, 11609 (2013).6. N. Shakhova et al., Nature Geoscience 7, 64 (2014) 7. R. E. Fisher et al., Geophys. Res. Lett. 38, L21803 (2011). See also R.E. Fisher et al., B31I-04, AGU Fall 2013 8. A. Biastoch et al., Geophys. Res. Lett., 38, L08602 (2011). 9. G. Whiteman et al., Nature 499, 401 (2013). See also equianos.com/wordpress/wpcontent/ uploads/Response-to-Whiteman et-al-Comment.pdf (2013). 10. P. Bergamaschi et al., J. Geophys. Res. 118, 7350 (2013). 11. BP Statistical Review of World Energy June 2013, see bp.com/statisticalreview. 12. A. Karion et al., Geophys. Res. Lett. 40, 4393 (2013).

Acknowledgements: Supported in part by the UK Natural Environment Research Council MAMM and Tropical Methane projects, the European Union's Ingos project, and Royal Holloway. Panel A prepared by EJD; Panel B by K. Masarie at NOAA.

## Panel A

**Methane ups and downs.** Top: Globally averaged CH<sub>4</sub> mole fraction in nmol/mol (abbreviated ppb, in blue); deseasonalized trend curve is in red. Bottom: Growth rate of globally averaged methane calculated as time-derivative of trend curve above. Data accessible via <u>ftp://ftp.cmdl.noaa.gov/ccg/ch4/flask/event/</u>. Note the increase prior to 1992, the marked slowdown from 2000 to 2006, and resumed growth since 2007.

## Panel B

**Methane growth rate by latitude.** Contours of methane growth rate with sine of latitude, using US NOAA data. Blue contours (in ppb/year) show decline in atmospheric methane burden; warm colors show increasing methane. Dashed lines show polar circles and tropics. Plotting by sine latitude equally weights the results for surface area with latitude. NOAA zonal means available at <a href="http://www.esrl.noaa.gov/gmd/ccgg/mbl/">http://www.esrl.noaa.gov/gmd/ccgg/mbl/</a>

Dept. Earth Sciences, Royal Holloway, University of London, Egham TW20 0EX, UK.

US National Oceanic and Atmospheric Administration (NOAA), Earth System Research Laboratory, Boulder, CO 80305, USA.

Laboratoire des Sciences du Climat et de l'Environnement, CEA-CNRS-UVSQ,

Saclay, 91191, Gif-sur-Yvette, France