Blowin' in the wind

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It is increasingly important to quantify emissions of environmentally sensitive gases into the atmosphere. **Bill Hirst**, **Philip Jonathan**, **David Randell** and colleagues[1] show that provided you understand how the atmosphere mixes as it moves, the answer to "who is emitting what, how much and where" is literally blowing in the wind.

Bob Dylan probably was not thinking of atmospheric gas dispersion when he wrote these words: but he was nonetheless right. Whether you are interested in monitoring greenhouse gas emissions, locating airborne viruses, or just finding a mate by detecting individual pheromone molecules – as moths do – then understanding gas dispersion is what you have to do.

Leaving the moths to their own devices, the issue of tracking down, mapping and monitoring sources of greenhouse gases – and in particular of methane – is pressing. Methane is one of the most significant of greenhouse gases. Atmospheric methane is responsible for about one third of the global warming effect of carbon dioxide, despite its concentration being less than one two-hundredth that of carbon dioxide. There is a strong incentive to reduce methane emissions to the atmosphere, and to do so we need to know how much is being emitted and from where. Wetlands account for around one fifth of global methane emissions. The energy industry, animal waste, and changes in land use are other important sources. Landfills are prodigious sources of methane – about one quarter of the United States' man-made methane emissions are from landfills: they are the largest single source of anthropogenic methane [2]. One way of comprehensively monitoring these and other emissions would be by airborne sensing.

What substances such as viruses, methane, smoke and pheromones have in common is that at low concentrations they are passively transported – they are just carried along in the air – and their path reflects the average wind speed and direction. Less obviously, the rate at which they spread out and dilute is governed by atmospheric turbulence. Imagine observing a plume of smoke from a smoke stack. When the ratio of turbulence to wind speed is low – during the night, say – the plume is long and thin. But during the day, thermals increase atmospheric turbulence and the plume spreads out. Combine this understanding with the constraint of mass continuity and you see that the further away from a source, the wider a gas plume will be and the lower its average gas concentration. At any downwind location, the mass of substance traversing the plume cross-section per second must equal the mass emitted per second by the source. This behaviour is captured in what is called a Gaussian plume eddy diffusivity model. It is a simple way of describing air movement and mixing based on a few directly measurable parameters[3].

Advanced optical gas sensors can rapidly and precisely measure concentrations down to parts per billion (ppb) levels using laser-diode absorption, effectively "counting" the number of molecules present. Mounted on an aircraft, these sensors can be used to measure concentrations along the flight path at numerous locations downwind of a source. Then all you need is a friendly statistician to infer the location and strength of the emission sources, along with the naturally occurring atmospheric background concentration that will be present.

We model the atmospheric concentration of methane at a remote location as the sum of the methane that is already in the air – the background concentration – and emissions from the ground that are propagated in the wind. The Gaussian plume eddy diffusivity model describes wind propagation using just five parameters: wind

Survey aircraft. Image reproduced courtesy of Sander Geophysics Ltd

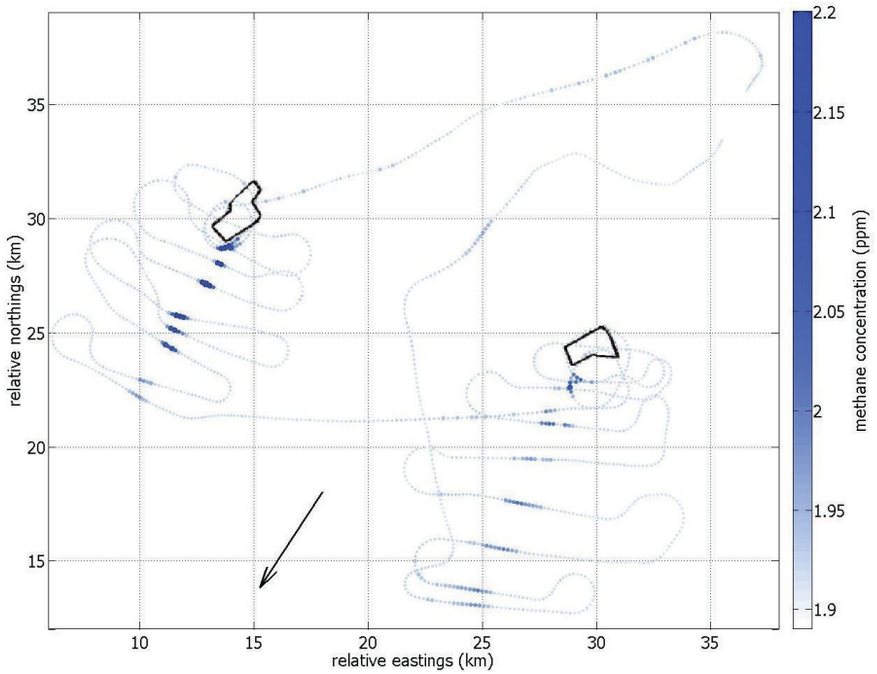

Figure 1. Flight trajectory around two landfills. Blue marker size and colour saturation indicate strength and location of measured excess methane concentrations. Arrow shows average direction of predicted air movement during flight. Polygons show perimeters of methane-emitting landfill areas. The aircraft takes off in the north-east corner and flies over the left-hand landfill first. The greater concentration of methane downwind of the landfill sites can be clearly seen

speed and direction, turbulence measures for horizontal and vertical directions, and the height of the atmospheric boundary layer – which we shall come to later. All these parameters can be measured or estimated in space and time for the region and duration of the flight. Adopting this simple wind propagation model has several advantages. In particular, we can treat the values of its parameters as measured with error, and then infer their bias and uncertainty as part of the modelling. We also know – from field measurements – that background concentration typically varies slowly in both space and time, whereas emission-related concentrations vary quickly. This difference is critical to our ability to distinguish the two in the model.

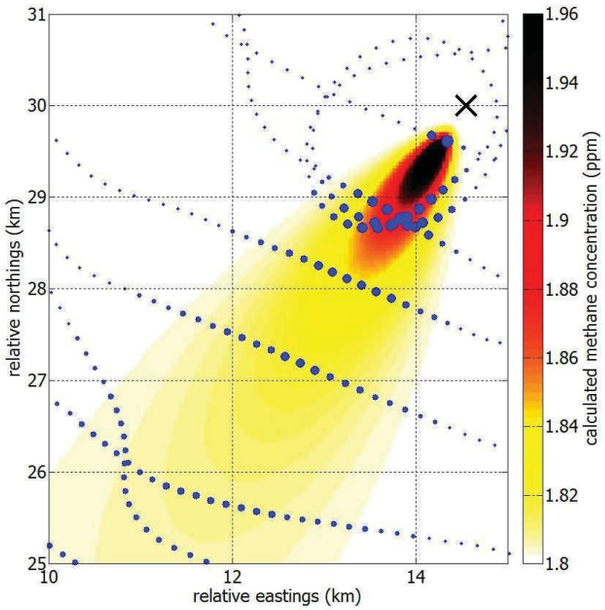

Figure 2. For comparison, a theoretical distribution of methane from the western landfill site. Calculated for an altitude of 200 m from a notional source (marked with a cross) at the centre of the western landfill emitting at 0.2 $m^3 s^{-1}$ of pure gas, assuming a background concentration of 1.8 ppm and actual wind conditions. Concentrations calculated along the flight trajectory shown as blue dots, with size proportional to excess concentration

Suppose we wish to quantify methane emissions from a landfill. The global average atmospheric methane concentration is approximately 1.8 parts per million (ppm, or 1800 ppb) and is increasing at 0.008 ppm per annum. Background methane concentration can vary by up to 0.02 ppm per day, due to changes in the depth of the atmospheric boundary; this is the layer of the atmosphere that contains all ground-level emissions for that day. Emissions accumulate near the ground during the night, when the atmospheric boundary layer is low, and are then diluted into the growing volume of the atmospheric boundary layer during the day, driven by the sun's warmth and the thermals that result. The top of the atmospheric boundary layer constitutes a "ceiling" on vertical transport of gases and particulates from the ground, and reduces the resulting rate of dilution downwind of a ground source. Clearly the aircraft must remain within the atmospheric boundary layer to detect concentrations from ground-level sources.

On a typical winter's night the atmospheric boundary layer is low – almost at ground level – and fogs and mists are prevalent. During a summer afternoon the atmospheric boundary layer can be up to 2 km high!

As part of testing our airborne sensing equipment and analysis method, we measured methane from two well-managed Canadian landfill locations using the aircraft trajectory illustrated in Figure 1. The aircraft flew at approximately 200 metres above ground level at around 50 metres per second ($m s^{-1}$), tracing a serpentine pattern up to 12 km downwind of each landfill. The ground domain we were interested in was approximately 40 km × 40 km. Referring to the figure, the aircraft took off from an airfield in the north-east and traversed the western landfill then the eastern before returning to the airfield. Concentrations measured along the flight trajectory are shown as blue dots, whose size is approximately proportional to "excess" concentration above background. The atmospheric boundary layer depth was approximately 400 m. Wind field data and supplementary meteorological data were obtained from the UK Meteorological Office. Wind speed was constant for the flight at 6.5 $m s^{-1}$ and wind direction approximately constant, as shown in the figure. As motivation, Figure 2 shows the concentrations that would

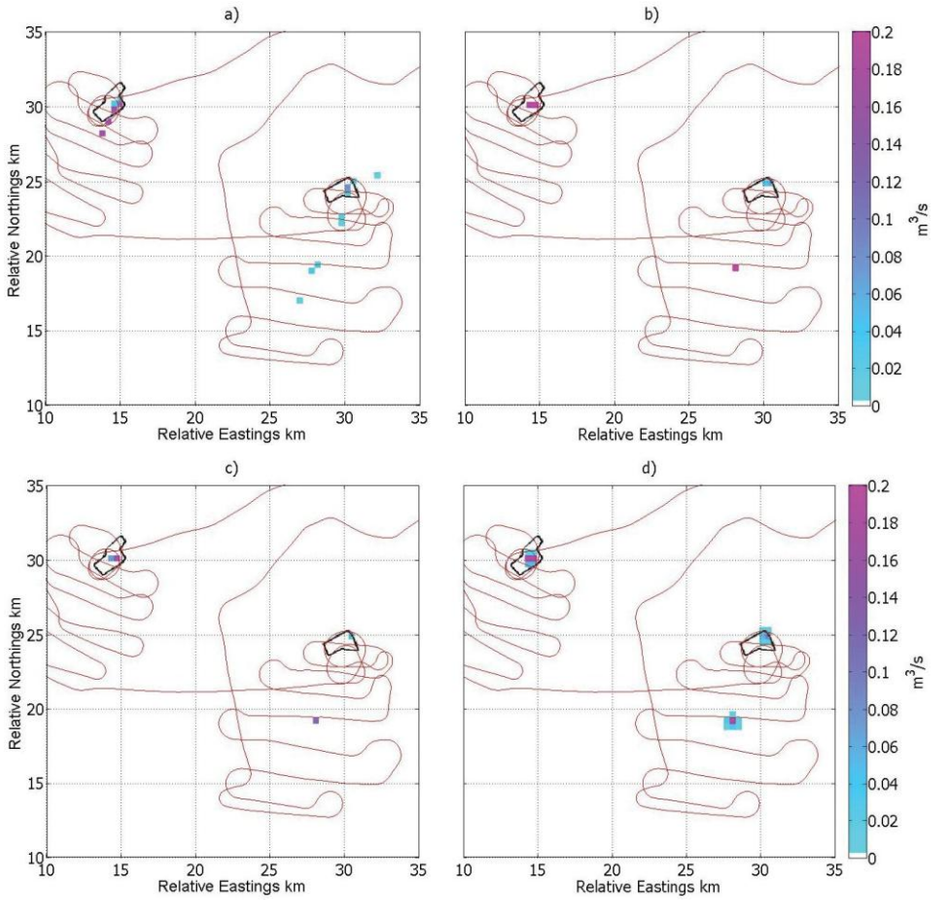

Figure 3. Estimated source emission rate maps for the landfill application. (a) Estimate from initial optimisation over a grid of potential source locations. (b) Median estimate from a mixture model estimated using reversible jump Markov chain Monte Carlo. (c) Marginal 2.5% credible value from the mixture model. (d) Marginal 97.5% credible value from the mixture model. Each panel shows a common sub-region of the original 40 km × 40 km domain (referenced with respect to the origin) within which all sources are estimated. Polygons indicate the perimeters of the landfills. The flight path is shown as a magenta line

arise at an altitude of 200 m from a known source of 0.2 cubic metres per second ($m^3 s^{-1}$) of pure gas placed at the centre of the western landfill emitting in identical wind conditions with a defined background concentration of 1.8 ppm – assuming our model to be correct!

Using the model we can also estimate the set of ground sources which best explain the actual measured airborne concentrations, as shown in Figure 3. We start by finding a good starting-point solution, shown in Figure 3(a). In addition to sources located within the landfills, some spurious sources appear downwind of both landfills. We believe these are due to changes in the wind field during the extended transit times from source to measurement locations up to 12 km away. In reality, there are many possible sets of ground sources which agree well with the measured concentrations. We therefore use Bayesian inference[4,5] to estimate the whole distribution of possible sets of ground sources which explain our measurements well. Figure 3(b) shows the estimated median source emission rate. Sources are centred within the landfills with just a single spurious source downwind of the eastern landfill. Interestingly, a 2 degree wind field bias correction was estimated which perhaps explains the improved solution. We summarise source uncertainty in term of 2.5% and 97.5% credible values for source emission rates, shown in Figure 3(c) and (d); no other spurious sources appear, suggesting strong spatial localisation of sources.

We have conducted several other measurement campaigns, including emissions from an industrial flare stack at a North African coastal location. The statistician on the team proved sufficiently persuasive and influential – for once – to achieve a reasonably well-designed experimental aircraft trajectory of eight multi-looped circuits of the flare stack at different altitudes and distances. A wind direction bias correction (of around 18 degrees) proved important, since estimation of coastal wind fields is particularly difficult – especially late in the day, which was when this opportunity for experimentation occurred. Current work is focused on adapting our approach to ground-based line-of-sight path-integrated gas concentration measurement: gas molecules in the path of a laser beam absorb some of the laser light, enabling average gas concentration along the path to be measured. These techniques are particularly well suited to measuring emissions from smaller defined areas, such as landfills, process plant and onshore carbon storage sites. Experimental design ideas are vital to optimise layouts of laser source sensors and retro-reflectors within the local constraints imposed by site characteristics and historic wind data.

There are numerous opportunities for development of airborne sensing, both from the sensor and modelling perspectives. We are working on modelling multiple flights and intermittent sources, and on incorporating measurements from complementary sensor technologies. Methane may be blowin' in the wind for some time to come, but at least we shall be able to tell where it's coming from!

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References

1. Hirst, B., Jonathan, P., Randell, D., González del Cueto, F. and Kosut, O. (2013) Locating and quantifying gas emission sources using remotely obtained concentration data. *Atmospheric Environment*, **74**, 141–158.

2. International Energy Agency (2009) *Energy Sector Methane Recovery and Use: The Importance of Policy*. P `http://www.iea.org/papers/2009/methane_brochure.pdf`

3. Gifford, F. A. (1976) Atmospheric dispersion models for environmental pollution. In D. A. Haigen (ed.), *Lectures on Air Pollution and Environmental Impact Analysis*. Boston: American Meteorological Society.

4. Gammermann, D. and Lopes, H. F. (2006) *Markov Chain Monte Carlo: Statistical Simulation for Bayesian Inference*. Boca Raton, FL: Chapman & Hall/CRC.

5. Green, P. J. (1995) Reversible jump Markov chain Monte Carlo computation and Bayesian model determination. *Biometrika*, **82**, 711–732.

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