ATMOSPHERIC CHEMISTRY

The return of ethane

Ethane emissions can lead to ozone pollution. Measurements at 49 sites show that long-declining atmospheric ethane concentrations started rising in 2010 in the Northern Hemisphere, largely due to greater oil and gas production in the USA.

Hannele Hakola and Heidi Hellén

Exposure to high concentrations of ground-level ozone can trigger asthma or weakening of lung function, and causes considerable damage to vegetation. Ozone is formed in the atmosphere from reactions involving nitrogen oxides and non-methane hydrocarbons, such as ethane, in the presence of sunlight. Ozone levels can increase when emissions of these compounds are high, temperatures are warm, and the sun is shining. The way to control ozone concentrations is to reduce the emissions of the precursor gases, including ethane. From 1984 to 2010, ethane emissions decreased from 14.3 to 11.3 terragrams of ethane per year, mainly as a result of reduced fugitive fossil fuel emissions[1]. However, writing in *Nature Geoscience*, Helmig *et al.*[2] present measurements of atmospheric ethane from a global observation network that indicate that the declining trend stopped and reversed in the Northern Hemisphere between 2009 and 2014.

Ethane, which is the longest-lived non-methane hydrocarbon, is emitted naturally from a variety of sources, including volcanoes, biomass burning and fossil carbon deposits. But anthropogenic activity, particularly related to fossil fuel extraction and distribution, became an increasingly important source of ethane during the twentieth century (Fig. 1). Atmospheric ethane concentrations, which are now roughly three times larger than pre-industrial levels, increased throughout most of the twentieth century and reached a peak around 1970. But then, starting in the 1970s, the implementation of stricter policies for limiting emissions of pollutants and pollutant precursors led to a gradual decline in atmospheric non-methane hydrocarbon concentrations, which continued through the first decade of the twenty-first century.

Helmig and colleagues[2] evaluated ten years of weekly to bi-weekly measurements of ethane and propane concentrations at 44 remote stations that are part of the National Oceanic and Atmospheric Administration's (NOAA) Global Greenhouse Gas Reference Network. They also used *in situ* measurements from five stations where samples are collected continuously or once a day. These data revealed that ethane concentrations have increased by 2.9–4.7% yr^{-1} in the Northern Hemisphere since 2009, with much smaller increases in the Southern Hemisphere.

Figure 1 | Natural gas flares in the Bakken shale formation. Atmospheric concentrations of ethane, a non-methane hydrocarbon, had been declining since the 1970s following the institution of air quality control policies. Using a long-term global network of ethane and propane observations, Helmig *et al.*[2] show that this declining trend reversed in 2009 in the Northern Hemisphere, probably due to increased emissions from oil and gas production in North America.

Support for the reversal of the long-term decline in ethane concentrations was found by Helmig *et al.* in ground-based atmospheric column measurements of ethane from Jungfraujoch, Switzerland, which provide a good representation of the continental background concentrations of ethane in the free troposphere. These measurements described a slight decline in Northern Hemisphere ethane concentrations from 1995 to 2009, but concentrations increased by 4.2% yr^{-1} between 2009 and 2014. In contrast, no trend reversal was evident in column measurements from New Zealand. A similar change in methane and ethane trends has also been observed in column measurements from Zugspitze, Germany[3].

In addition to investigating the trends in atmospheric concentrations, Helmig *et al.* looked for the sources that were responsible for the trend reversal. They used propane, which only persists in the atmosphere for about a quarter as long as ethane and is co-emitted from most ethane sources, to help identify the potential sources of additional ethane emissions. Since propane has a shorter lifetime, it is transported over shorter distances in the atmosphere than ethane, and it is therefore only detected within relatively close proximity to its emissions source. Helmig and co-workers find that propane has increased substantially only in the USA and downwind areas, whereas propane concentrations in central Europe, the Pacific region and the Southern Hemisphere have been relatively stable. These findings suggest that the increase in ethane concentrations is due to emissions associated with a substantial increase in oil and natural gas production in the USA. However, one of the highest increases in propane concentration was observed in Tiksi, Russia, which suggests that North America is not the sole source of increased emissions.

Ethane is also co-emitted from oil and natural gas sources with methane, a powerful greenhouse gas. Time-series measurements of methane and ethane were significantly correlated during 2007–2014 (ref. 3), a period when global atmospheric methane concentrations began increasing again after stalling between 1999 and 2007. However, there was no correlation between ethane and methane concentrations in measurements earlier than 2007, or in measurements from the Southern Hemisphere, suggesting that oil and gas production have become more important sources of methane in the Northern Hemisphere since 2007. Based on the ethane-to-methane ratio of emissions and assuming that the increase in ethane emissions is entirely from oil and natural gas sources, these sources contributed at least 39% to the renewed methane increase.

Measurements of ethane emissions from the Bakken shale formation in North Dakota support the notion that North American oil and gas production are contributors to the Northern Hemisphere ethane trend[4]. Oil production in the formation has increased by a factor of 3,500 over the last decade, and gas production has increased by a factor of 180. To evaluate how this production increase is affecting ethane emissions, continuous airborne measurements of ethane were made over the North Dakota portion of the Bakken shale formation in spring 2014 (ref. 4). If the emissions measured during the airborne study are representative, the formation is responsible for emissions on the order of 0.23 ± 0.07 terragrams of ethane per year, equivalent to 1–3% of all ethane emissions globally[4].

In order to understand some of the air quality impacts of these large increases in ethane, Helmig *et al.* modelled the effect of observed non-methane hydrocarbon increases on ozone formation. The increase in emissions estimated by Helmig *et al.* could result in additional ozone formation of 0.5 nmol mol^{-1} yr^{-1} for June to August in areas downwind of oil and gas production sites in North America. There is a danger that these non-methane hydrocarbon emission changes can offset emission policies and controls aimed at reducing ozone concentrations.

Combined with measurements of ethane that are representative of the free troposphere[3] and a large oil and gas field[4], the work by Helmig *et al.*[2] shows that ethane and propane concentrations in the Northern Hemisphere have taken an upward turn, largely as a by-product of the massive increase in oil and gas exploration in North America. These oil and gas operations are threatening to reverse what had been an important success story: decades of declining air pollution in North America. ❐

Hannele Hakola and Heidi Hellén are at the Finnish Meteorological Institute, PL 503, FIN-00101 Helsinki, Finland.
e-mail: hannele.hakola@fmi.fi

References

1. Helmig, D. *et al. Nature Geosci.* http://dx.doi.org/10.1038/ngeo2721 (2016).
2. Simpson, I. J. *et al. Nature* **488,** 490–494 (2012).
3. Hausmann, P., Sussmann, R. & Smale, D. *Atmos. Chem. Phys.* **16,** 3227–3244 (2016).
4. Kort, E. A. *et al. Geophys. Res. Lett.* http://doi.org/bhf2 (2016).

Published online: 13 June 2016