

## **Renewed methane increase (2007–2014): contribution of oil and natural gas emissions determined from methane and ethane column observations**

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Harmonized time series of column-averaged mole fractions of atmospheric methane and ethane over the period 1999–2014 are derived from solar Fourier transform infrared (FTIR) measurements at the Zugspitze summit (47° N, 2964 m a.s.l.) and at Lauder (45° S, 370 m a.s.l.). Long-term trend analysis reveals a consistent renewed methane increase since 2007 of 6.2 [5.6, 6.9] ppb yr<sup>-1</sup> at the Zugspitze and 6.0 [5.3, 6.7] ppb yr<sup>-1</sup> at Lauder (95 % confidence intervals). Several recent studies provide pieces of evidence that the renewed methane increase is most likely driven by two main factors: (i) increased methane emissions from tropical wetlands, followed by (ii) increased thermogenic methane emissions due to growing oil and natural gas production. Here, we quantify the magnitude of the second class of sources, using long-term measurements of atmospheric ethane as tracer for thermogenic methane emissions. In 2007, after years of weak decline, the Zugspitze ethane time series shows the sudden onset of a significant positive trend ( $2.3 [1.8, 2.8] \times 10^{-2}$  ppb yr<sup>-1</sup> for 2007–2014), while a negative trend persists at Lauder after 2007 ( $-0.4 [-0.6, -0.1] \times 10^{-2}$  ppb yr<sup>-1</sup>). Zugspitze methane and ethane time series are significantly correlated for the period 2007–2014 and can be assigned to thermogenic methane emissions with an ethane-to-methane ratio of 10–21 %. We present optimized emission scenarios for 2007–2014 derived from an atmospheric two-box model. From our trend observations we infer a total ethane emission increase over the period 2007–2014 from oil and natural gas sources of 1–11 Tg yr<sup>-1</sup> along with an overall methane emission increase of 24–45 Tg yr<sup>-1</sup>. Based on these results, the oil and natural gas emission contribution  $C$  to the renewed methane increase is deduced using three different emission scenarios with dedicated ranges of methane-to-ethane ratios (MER). Reference scenario 1 assumes an oil and gas emission combination with MER = 3.3–7.6, which results in a minimum contribution  $C > 28$  % (given as lower bound of 99 % confidence interval). For the limiting cases of pure oil-related emissions with MER = 1.7–3.3 (scenario 2) and pure natural gas sources with MER = 7.6–12.1 (scenario 3) the results are  $C > 13$  % and  $C > 53$  %, respectively. Our results suggest that long-term observations of column-averaged ethane provide a valuable constraint on the source attribution of methane emission changes and provide basic knowledge for developing effective climate change mitigation strategies.