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UK landfill methane emissions: Use of mobile plume measurements and carbon isotopic characterisation to reassess oxidation rates for open and closed sites

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Biological methane oxidation in landfill cover material can be characterised using stable isotopes. Methane oxidation fraction is calculated from the carbon isotopic signature of emitted CH_4, with enhanced microbial consumption of methane in the aerobic portion of the landfill cover indicated by a shift to less depleted isotopic values in the residual methane emitted to air. This study was performed at four southwest England landfill sites. Mobile mole fraction measurement at the four sites was coupled with Flexfoil bag sampling of air for high-precision isotope analysis. Gas well samples collected from the pipeline systems and downwind plume air samples were utilized to estimate methane oxidation rate for whole sites. This work was designed to assess the impact on carbon isotopic signature and oxidation rate as UK landfill practice and waste streams have changed in recent years.

The landfill status such as closed and active, seasonal variation, cap stripping and site closure impact on landfill isotopic signature and oxidation rate were evaluated. The isotopic signature of ^{13}C-CH_4 values of emissions varied between -60 and -54‰, with an averaged value of -57 +- 2‰ for methane from closed and active landfill sites. Methane emissions from older, closed landfill sites were typically more enriched in ^{13}C than emissions from active sites. This study found that the isotopic signature of ^{13}C-CH_4 of fugitive methane did not show a seasonal trend, and there was no plume observed from a partial cap stripping process to assess changes in ^{13}C-CH_4 isotopic signatures of emitted methane. Also, the closure of an active landfill cell caused a significant decrease in mole fraction of measured CH_4, which was less depleted ^{13}C in the emitted plume due to a higher oxidation rate. Methane oxidation, estimated from the isotope fractionation, ranged from 3 to 27%, with mean values of 7% and 15% for active and closed landfills, respectively. These results indicate that the oxidation rate is highly site specific.