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Integral emission factors for methane determined using urban flux measurements and local-scale inverse models

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The most important long-lived greenhouse gas (LLGHG) emitted during combustion of fuels is carbon dioxide (CO_2), however also traces of the LLGHGs methane (CH_4) and nitrous oxide (N_2O) are released, the quantities of which depend largely on the conditions of the combustion process. Emission factors determine the mass of LL-GHGs emitted per energy used (or kilometre driven for cars) and are key inputs for bottom-up emission modelling. Emission factors for CH_4 are typically determined in the laboratory or on a test stand for a given combustion system using a small number of samples (vehicles, furnaces), yet associated with larger uncertainties when scaled to entire fleets. We propose an alternative, different approach - Can integrated emission factors be independently determined using direct micrometeorological flux measurements over an urban surface? If so, do emission factors determined from flux measurements (top-down) agree with up-scaled emission factors of relevant combustion systems (heating, vehicles) in the source area of the flux measurement?

Direct flux measurements of CH_4 were carried out between February and May, 2012 over a relatively densely populated, urban surface in Vancouver, Canada by means of eddy covariance (EC). The EC-system consisted of an ultrasonic anemometer (CSAT-3, Campbell Scientific Inc.) and two open-path infrared gas analyzers (Li7500 and Li7700, Licor Inc.) on a tower at 30m above the surface. The source area of the EC system is characterised by a relative homogeneous morphometry (5.3m average building height), but spatially and temporally varying emission sources, including two major intersecting arterial roads (70.000 cars drive through the 50% source area per day) and seasonal heating in predominantly single-family houses (natural gas). An inverse dispersion model (turbulent source area model), validated against large eddy simulations (LES) of the urban roughness sublayer, allows the determination of the spatial area that contributes to each measurement interval (30 min), which varies with wind direction and stability. A detailed geographic information system of the urban surface combined with traffic counts and building energy models makes it possible to statistically relate fluxes to vehicle density (km driven) and buildings (gas heated volume) - and ultimately quantify the contribution of space heating, transport sector and fugitive emissions to the total emitted CH_4 from an urban environment.

The measured fluxes of CH_4 over the selected urban environment averaged to 22.8 $\mathrm{mg\ CH_4\ m^{-2}\ day^{-1}}$ during the study period. Compared with the simultaneously measured CO_2 emissions, the contribution of CH_4, however, accounts for only about 3% of the total LLGHG emissions from this particular urban surface. Traffic contributed 8.8 $\mathrm{mg\ CH_4\ m^{-2}\ day^{-1}}$, equivalent to 39% of the total CH_4 flux. The determined emission factor for the typical fleet composition is 0.062 $\mathrm{g\ CH_4}$ per km driven which is higher than upscaled fleet emission factors (EPA) by a factor of two. This discrepancy can be partially explained through the slower city traffic with frequent idling (traffic congestion), fleet composition and cold starts. Emissions of CH_4 by domestic space heating (55% of the total CH_4 flux or 12.7 $\mathrm{mg\ CH_4\ m^{-2}\ day^{-1}}$) are also higher than estimated from upscaled emission factors. There is no evidence of substantial unknown sources such as soil processes, combustion of wood, and leakages from gas distribution pipes (residual: 6% or 1.3 $\mathrm{mg\ CH_4\ m^{-2}\ day^{-1}}$). The presented study is among the first direct measurements of CH_4 emissions over an urban surface and demonstrates that flux measurements of greenhouse gases can be used to determine sources and emission factors in complex urban situations.