**FTIR**

**Quantification of methane emissions in Hamburg using a network of**

**FTIR spectrometers and an inverse modeling approach**

* Total Colom measument with a computational fluid dynamics (CFD) mode
* At a city scale, the mix of sources can however be quite complex and also above ground-level sources, which cannot be picked up very well using ground-based mobile surveys, can play a role in the total emissions
* The EM27/SUN is an instrument commonly used to measure   
  column-averaged dry air mole fractions of CH 4 with high precision
* The network of solar-tracking spectrometers measures the total column concentration of CH 4 and   
  thus is sensitive to both near ground and above ground sources.
* When quantifying CH 4 emissions, usually only one of the concepts of mobile measurements or inversion of column/in-situ  
   measurements is applied. In this study we combined both of them to identify and quantify the sources in a top-down approach
* We used a sensor network similar to MUCCnet together with an emission map with updated distributions based on mobile   
  in situ measurements at the street level.
* The TNO GHGco Inventory is an European database   
   that includes spatially resolved emission data for CO 2 , CH 4 , CO, NO x and NMVOCs
* Before and after that, side-by-side measurements of the   
  four spectrometers were carried out on a roof at the University of Hamburg to make sure all instruments are properly calibrated   
  to each other.
* The EM27/SUN instruments were deployed in custom enclosures that protect the spectrometer from rainfall and adverse   
  weather conditions (Dietrich et al. (2021); Heinle and Chen (2018)).
* Between the 27th of July 2021 and the 9th of Sept 2021 our four FTIR-Spectrometers were measuring in Hamburg.
* n August 2021 the weather was unexpectedly cloudy and many days the systems were   
  in idle. However, we still had nine good measurement days with sufficient sun-shine to carry out the measurements
* The measurements of the column-averaged dry air mole fractions must be properly filtered to exclude measurement errors.   
  In particular, these arise from non-optimal solar tracking, which is mainly caused by clouds. We used two successive filtering 125   
   steps: The first filtering step is based on physical properties such as solar elevation, absolute solar intensity, and solar intensity   
  variation during a Michelson interferometer scan. The second filtering step uses data statistics to remove outliers and measure-  
  ment periods with too few data points. In addition, the measurement data are averaged using a 10 minutes moving average   
  filter.
* In order to filter out days with fragmented and interrupted measurements due to repeated cloud cover, we only consider 130   
   measurement days when at least two stations were measuring at the same time for more than 5 hours
* To support the modelling and the calculation of the final emission estimate, in situ measurements were performed with a Picarro   
  GasScouter G4302 which measures CH 4 and C 2 H 6 and a Picarro G2301 greenhouse gas analyzer which measures CH 4 and   
  CO 2
* We deployed an 190   
   Isotope Ratio Mass Spectrometer (IRMS) which continuously measures δ13C and δD with a Delta V Plus/Deltaplus XL from   
  Thermo Fisher Scientific Inc

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* In order to quantify the urban emissions based on the concentration measurements, a Bayesian inversion framework was used.
* n order to generate those backward trajectories and the footprints, the STILT (Stochastic Time-Inverted Lagrangian Trans-220   
   port) model is used
* The meteorological input data for this model is provided by the ERA5 data-set (Muñoz Sabater (2019)).   
  As prior information, the TNO GHGco inventory is used (Super et al. (2020)).
* The correlation increased significantly when including the natural source into the modeling as visible in Figure A2 in 295   
   Appendix.

Differential column measurements using compact   
solar-tracking spectrometers

* differential column   
  measurement has a precision of 0.01 % for XCO2 and XCH4

Assessing Urban Methane Emissions using Column Observing   
Portable FTIR Spectrometers and a Novel Bayesian Inversion   
Framework

**MUCCnet: Munich Urban Carbon Column network**

* the permanent urban GHG net-  
  work in Munich, which is based on the differential column   
  measurement (DCM) principle and consists of five fully au-  
  tomated FTIR spectrometers
* Column measurements have proven to be a powerful tool   
  for assessing GHG emissions from cities and local sources,   
  because they are relatively insensitive to the dynamics of the   
  boundary layer height and surface fluxes upwind of the city if   
  a differential approach is used
* he column-averaged   
  concentrations of a gas in the atmosphere are measured up-  
  wind and downwind of an emission source, utilizing ground-  
  based FTIR spectrometers that use the sun as a light source.  
  The concentration enhancements between the two stations   
  are caused by the urban emissions
* After about   
  5 d, when the a priori vertical pressure profiles from NCEP   
  (National Centers for Environmental Prediction) are avail-  
  able, the retrieval algorithm converts the information from   
  the interferograms into concentrations. The retrieval algo-  
  rithm used is GGG2014 (Wunch et al., 2015), which is also   
  used to retrieve all the TCCON data. We applied the standard   
  TCCON parameters, including the air-mass-independent cor-  
  rection factors. The spectral windows for retrieving diverse   
  gas species are slightly modified according to the EGI setup   
  (Hedelius et al., 2016).

**The Differential Colum Measurement**

A Fourier transform Infrared Spectrometer network has been deployed inside and around the city borders to investigate the urban greenhouse gas (GHG) emissions from the city region of Hamburg. Parts of he usually in Munich located sensor network MUCnet has been relocated to Hamburg and was deployed for a shorter time period between the 01.08.2021 to the 31.9.2021. The network consists of four fully autonomous and automated Enclosures. These were located about 20 km from the city centre to the east, North and West, with one enclosure in the city close to the city centre at the Geomatikum in close proximity to the mass spectrometer. The location can be seen in Figure #####.

Figure ####

The enclosures operate by measuring the solar spectrum with a high temporal resolution of 90 seconds, which is in the revival process, later on, averaged over a 10 min time period to accurately calculate the CH\_4, CO\_2….. concentration in the total air column.

The enclosures are equipped with a Michelson interferometer (Bruker EM27/Sun). This spectrometer has an attached solar tracker that enables the Tracking of the Sun by redirecting the light rays with a set of electronically controlled gold-plated mirrors. The spectrometer is housed in a weatherproof aluminium box to protect it and its auxiliary equipment, like a computer, heating unit, control electronics, etc., from the elements. To enable an undisturbed light ray, the solar tracker sticks out the top of the box with an automated cover that opens at favourable weather conditions and aligns itself with the solar tracker. To protect the tracker from precipitation, a rain sensor and a cloud detection sensor are placed on top of the enclosure. They automatically initiate a closure of the cover when precipitation is detected or the cloud coverage is too great. The weatherprove enclosure and automated operation allow for autonomous data acquisition of a geographically extensive network with minimal staffing. Data collection during fluctuating weather conditions can be achieved as the measurement can be initiated and terminated quickly without lengthy setup times.

The significant advantage of the FTIR approach to other in situ measurements lies in the total column measurement, which measures the total air column of the atmosphere between the light emitter, i.e. the sun and the spectrometer. The spectral absorption by the methane can then accurately be measured for the entire atmosphere. Other approaches that measure concentration at the street level have the disadvantage that they can miss methane emission emitted above the instruments like high chimneys. The degree of mixing of the GHG with the background air is highly dependent on various airflow characteristics, Topography and Emission type and location. A total column measurement can’t negate all of them, but it gives a more reliable picture than simple in situ measurements.

A total column approach with solar spectroscopy has the added advantage of providing a reference for satellite measurements and vice versa.

The sensor network deployment achieves a differential total column measument, in which at least one sensor is always located upwind and one downwind of an emitter. The quantity of GHG released by an emitter can be estimated by comparing the concentration delta of the two columns.

A computational fluid dynamics (CFD) model is used to identify the Emission location. This is aided by using a Bayesian inversion framework.

An inverse Framework is a method where the effects of an event or condition are used to calculate the cause that leads to the observed effects. Here the methane concentration has been measured in the atmosphere, and it is attempted to calculate its emitters. In a forward method, the measured concentration in the air would be calculated from the known emitters.

To statistically improve the calculation of the cause, further prior information can be used. The TNO inventory, which has the highest spatial resolution of “known” emitters in Hamburg, is used in this case. This inventory is compiled by estimating emissions due to fossil fuel usage, density, type of emission, etc.

When applying the Basyan Approach in the inversion, the TNO GHGco inventory was used as the prior estimation of the emissions.

“The TNO GHGco Inventory is a European database that includes spatially resolved emission data for CO2, CH4, CO, NOx and NMVOCs, the spatial resolution is (1/60)◦ for longitude and (1/120)◦ for latitude” (Hamburg paper). This inventory is Hamburg's current highest resolution inventory, giving the best available prior emission estimation. Its limitation is the Bottom-up approach may not encounter all emission sources or does not yet account for relocated emitters and sporadic emitters.

In Bayes’s theorem, the posterior model is proportional to the prior model, which is the probability of the methane being emitted using previously acquired knowledge times the “likelihood of the Data”. The likelihood of the Data links the target model (the posterior of the measured variable) to the measured data. The likelihood is the probability of measuring a concentration given the emission from the Prior model.

* Formal: Posterior probability = (Prior probability \* Likelihood of the data )/ Normalization constant

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A Basyan inversion can estimate the methane emission of a source by using prior knowledge of the source by updating this knowledge with measurements of the Methane concentration in the air.

To identify the location, the inversion is also making use of the ###### weather model. This model is additionally corrected for the boundary layer height and wind direction by using a wind LIDAR. This corrected model is then used to generate backward trajectory footprints with a Stochastic Time-Inverted Lagrangian Transport (STILT) model. The Inversion is then able to use the STILT footprints for its identification of the Emission location.

**Bayses Theory**

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* Bayes’s theorem relates events or random variables by   
  avoiding using their joint probability in favor of conditional   
  and marginal ones. In many cases, these probabilities are   
  easier to define.  
  In mineral exploration, it provides the tools to update our   
  knowledge on the subsurface with data from different sources   
  of fuzzy information.  
  In geological modeling, Bayes’s theorem is used to trans-  
  form the prior distribution of the system’s parameters into a   
  narrower posterior distribution by using a set of uncertain   
  measurements of the response variables.
* Posterior probability = (Prior probability \* Likelihood of the data )/ Normalization constant
* Prior probability : P(A) Probability of event a, the probability bevor the measument due to prior knowlage
* Likelihood of the data: P(B|A) reliability of the survey (Probability that B given A Occurs
* Normalization constant: P(B) probability of a event, the measument made after the priory.
* Posterior probability: P(A|B) probability of A occoring given Event B. (Probability of existence.
* Bayesian inversion is a statistical method used in many geoscience applications to assess the probability distribution of the unknown variables conditioned on the available measurements, where the posterior model is uniquely defined by a prior model representing the knowledge of the variable before the data acquisition and a likelihood model linking the target variables and the measured data.
  + Postierior model is defend by the Prior model (the Knowlage before measuments) and the Liklyhood model (Linking the tagrgeng/prosteria variable to the measument)

**Inverse Model**

* An inverse problem in science is the process of calculating from a set of observations the causal factors that produced them
* It is called an inverse problem because it starts with the effects and then calculates the causes. It is the inverse of a forward problem, which starts with the causes and then calculates the effects.
* Very important in remote sensing