Remote Sensing of Emission Rates from Gaseous Emission Sources

Guest contribution

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Abstract

Fourier Transform Infrared (FTIR) spectrometry as a multi-component and path-averaging measurement method provides new information about emission sources. Effluent concentrations such as CO, CO_2, H_2O, NO, N_2O, NO_2, SO_2, and HCl from power plant smoke stacks, aircraft engines, and flares as well as the temperature of plumes are quantified from passive measurements with the spectra analysis software MAPS developed at the IFU. Using an infrared radiation source the upwind and downwind concentrations of CO, CO_2, NO, N_2O, CH_4, NH_3, H_2O, and HCHO near the ground are measured to determine the source (slurry spreading, composting windrows, road traffic, garbage deposits, open-cast lignite minings, an ensemble of small building smoke stacks, and forest fires) influence upon ambient air. With Gaussian dispersion theory and complementary on-site meteorological measurements the emission rates are calculated from these data.

Introduction

The knowledge of emission rates from gaseous emission sources is an important input information for chemical transport models. The emissions of diffuse sources and of sources which can not be determined by in situ methods are known very poor. Remote sensing is used to get new information about these emissions. The goal of the measurements at point sources (smoke stacks) and at diffuse, area and line sources (industry, traffic, agriculture, waste disposals) are the determination of diurnal variations of exhaust emissions and the evaluation of emission modelling (supported in frame of the program SANA by the German Ministry of Education, Science, Research, and Technology) as well as the determination of exhaust compounds from aircraft engines (supported by the German Research Foundation DFG) and flares (supported by Shell Research Limited).

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Instrumentation and measurement principles

To determine concentrations of effluents Fourier-Transform-Infrared (FTIR) spectroscopy can be performed for active and passive radiation measurements (emission and absorption spectroscopy). With this multi-component and path-averaging measurement method a large number of gases can be quantified simultaneously and cost-effectively [1]. The measurements were performed using a van equipped with the Kayser-Threde double pendulum interferometer K300. The spectrometer works at spectral resolutions of 0.06 to 10 cm^{-1} (typically 0.2 cm^{-1} for the measurements described here) in a spectral range of 700 to 7800 cm^{-1} depending on the detector configuration (Joule-Thomson cooled MCT or InSb detectors). A flat tracking mirror reflects radiation from the distant source into the spectrometer telescope with an aperture of 15 cm and a field of view of 3 mrad [2].

In the emission mode the radiation from the warm exhaust plumes are received from just above the stack top from distances up to 500 m with the mirror. This passive method enables unannounced measurements outside a plant. Exhausts of aircraft engines and flares are measured with the same configuration. The influence of atmospheric background radiation is eliminated by evaluating measurements with and without the plume. The foreground concentrations between the stack and the instrument can be obtained by active open-path FTIR absorption measurements using an infrared radiation source (globar).

To determine the trace gas concentrations in ambient air typical path lengths between 100 and 500 m were used. Two and more measurements are carried out perpendicular to the prevailing wind direction at several downwind positions from the source in the whole effluent plume, thus obtaining path-averaged contributions of the heterogeneous releases across an area source. To determine the emission rate by inverse Gaussian dispersion modelling meteorological parameters such as temperature, humidity, pressure, global solar radiation, and the wind vector are determined near the absorption paths by complementary measurements.

Data interpretation

A multi-component air pollution software (MAPS) for the interpretation of infrared spectra has been developed at the IFU. It is based on a radiative transfer model using a line-by-line calculation for gaseous transmittances including the HITRAN line parameter data, Voigt line shapes, line overlaps, and continuum absorptions [3]. This procedure offers the possibility to include current atmospheric and geometrical parameters as well as spectroscopic features of the instrument (spectral resolution, line shape function) and to study interferences of spectral signatures of different compounds in gas mixtures. Spectral signatures

were investigated to define characteristic spectral regions (microwindows) for the single gases. The gas concentrations are calculated by least-squares fitting procedures (height or depth of spectral lines). Effects of multiple aerosol scattering as well as cloud and rain influence are included. The concentration retrieval is performed on-line to give the topical values of CO, CO_2, H_2O, CH_4, N_2O, NO, NO_2, SO_2, HCl, NH_3, HNO_3, and HCHO every 5 to 10 minutes. The detection limits for absorption measurements of greenhouse gases are below one tenth of their concentration in ambient air, and for other gases in the order of 1 to 10 ppb. Using spectral libraries about 100 VOC components can be identified at concentrations higher than 10 to 100 ppb.

In the emission mode the detection limits are in the order of 10 ppm. The mean deviation of the results in emission spectroscopy does rarely exceed ± 20 %, those of absorption measurements agree within ± 5 % compared to *in situ* measurements.

The emission rates of aligned sources are determined using mass-flow information from the plant staff [4]. In a test at a power station the streaming velocity in a chimney plume was measured with the cw-Doppler-lidar of the DLR giving mass fluxes with complementary concentration measurements by FTIR emission spectroscopy which differ by ± 25 % in relation to the fluxes of in situ measurements (CEM) of the plant staff [5].

The method to determine emission rates of diffuse sources with Gaussian modelling was evaluated by measuring the CO emissions from road traffic on a highway during constant conditions (about 30 cars/minute). Several absorption paths of 250 m length parallel to the highway were used at distances of 30 to 500 m to the highway. The calculated emission rates varied between 151 and 163 mg/(m^2 s), *e.g.* were invariant from the distance of measurements to the highway [6].

Measurement results

Aircraft exhausts (CO, NO, CO_2, H_2O)

After finding out spectral regions in which the IR-active compounds of the exhaust show clear features and developing a multi-layer retrieval software based on MAPS different aircraft engines (GE CF700-2D2, RR M45H, JT8D-15, CFM56-3B, CFM56-5C2) were investigated. The detection limits for a typical exhaust plume (ϕ = 50 cm, T = 380° C) were found to be 20 ppm for CO, 90 ppm for NO, 100 ppm for CO_2, and 0.7 % for H_2O. The method was validated with exhaust sample analyses of a gas burner (± 20 % deviation) [7].

Pig-farming **(NH_3, CH_4, N_2O, CO_2, NO)**

FTIR measurements at different absorption pathes were performed inside the stalls of pig farming. Emission rates were calculated using the ventilator throughputs. The ammonia emission rates from different stalls vary between 8.76 and 3.24 kg/(pig·year) [8]. The emission model from the IER of the University Stuttgart gives a mean value of 5 kg/(pig·year).

Slurry spreading **(NH_3)**

The ammonia emission rates were measured during and after spreading of 160000 l slurry on grassland of an area 150 m wide and 150 m long, which 7.5 hours later was ploughed mechanically. The maximum concentration of 410 ppb (emission rate 0.056 mg/(m^2 s)) was detected at the end of the spreading followed by an almost exponential decrease down to 200 ppb (0.015 mg/(m^2s)) seven hours later. After ploughing the concentration was found to be 80 ppb [6].

Highways **(CO, NO, CH_4, VOC)**

During a mean car density of 45 cars/minute a CO emission rate of 0.34 mg/(m^2 s) and a total emission of 6.8 g/s were measured [6]. The emission modelling by the IER of Universität Stuttgart for the same conditions gave 6.9 g/s.

References

1. R. Haus, K. Schäfer; W. Bautzer, J. Heland, H. Mosebach, H. Bittner, T. Eisenmann; *Appl. Opt.* 33 (1994) 5682-5689.
2. H. Mosebach, T. Eisenmann, Y. Schulz-Spahr, I. Neureither, H. Bittner, H. Rippel, K. Schäfer, D. Wehner, R. Haus; Remote Sensing of Smoke Stack Emissions Using a Mobile Environmental Laboratory. *Proc. SPIE, Vol.* **1717**, Environmental Sensing, Berlin 1992, pp. 149-158.
3. K. Schäfer, J. Heland; Multicompnent Air Pollution Software (MAPS). *Software User Guide*, Fraunhofer-Institut für Atmosphärische Umweltforschung, Garmisch-Partenkirchen (1996).
4. R. Haus, K. Schäfer, J.I. Hughes, J. Heland, W. Bautzer; FTIS remote sensing of smoke stack and test flare emissions. in: P. Fabian, V. Klein, M. Tacke, K. Weber, C. Werner (eds.), *Proc. SPIE, Vol.* **2506**, Air Pollution and Visibility Measurements. Munich 1995, pp. 45-54.
5. C. Werner, R. Haus, F. Köpp, K. Schäfer; *Appl. Phys.* **B 61** (1995) 249-251.

6. R. Haus, K. Schäfer, W. Bautzer; Emission rate determination for air pollutants in agriculture and urban areas based on open-path FTIS measurements. in: P. Fabian, V. Klein, M. Tacke, K. Weber, C. Werner (eds.), *Proc. SPIE,*. **2506,** Air Pollution and Visibility Measurements. Munich 1995, pp. 247-256.

7. J. Heland, K. Schäfer, R. Haus; Remote sensing and gas analysis of aircraft exhausts using FTIR emission spectroscopy. in: P. Fabian, V. Klein, M. Tacke, K. Weber, C. Werner (eds.), *Proc. SPIE,* **2506,** Air Pollution and Visibility Measurements. Munich 1995, pp. 62-71.

8. R. Haus, K. Schäfer, J. Heland, C. Salow, J. Sciborski; Stock-Farming, Agriculture, and Waste Emission Control Based on Open-Path FTIS and in situ Measurements. In: 87th Annual Meeting and Exhibition of the Air and Waste Management Association, 19.-24.06.1994, Cincinnati, *Conf. Proc., Air and Waste Management Association,* Pittsburgh 1994.