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Determination of continuous trace gas distribution maps for e.g. NO2, O3 and SO2 in a city using tomographic LP-DOAS measurements

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Measurements of trace gases are nowadays performed in all larger cities for urban air pollution monitoring by applying few in-situ measurement stations. The concentrations of many trace gases in urban areas mainly depend on anthropogenic emissions, primarily from industry, cars and heating systems but also depend on geographical conditions and local transport processes. Thus the real concentration may vary strongly on scales of several 10 to 100m which are not captured by these standard measurements. But the knowledge of the exact concentration and their distribution on regional or local scale is necessary for air pollution monitoring, to estimate emission sources and sinks and their strength, to study chemical transformation processes and to validate chemical transport models and provide realistic input data for such models. However measurements of trace gas distributions with temporal resolution of minutes are not possible with available standard measurement methods.

Here we present tomographic Long path Differential Optical Absorption Spectroscopy (LP-DOAS) measurements which allow deriving two and three dimensional distributions of trace gases by measuring the average concentration along 10 to 20 intersecting light paths and applying tomographic inversion techniques. We developed and applied such a setup for the first time to derive the horizontal distribution of several trace gases in the open atmosphere. The presented measurements took place in the city of Heidelberg, Germany from 2005 to 2007 and focused on the trace gases NO2, SO2, O3, HCHO and HONO, which play a major role in the polluted atmosphere. The setup consisted of three Multi Beam LP-DOAS instruments and 20 retro reflector arrays all installed on different buildings in the city. The 20 realised intersecting light paths covered an area of 4×4 km^2 with different emission sources.

We developed an optimised tomographic retrieval for such measurement data by applying the algebraic reconstruction method SIRT (Simultaneous Iterative Reconstruction Technique). We show that optimising the reconstruction parameters for the applied setup may have a significant influence on the derived results and errors. With the found best parameters we could retrieve long term time series of horizontal trace gas distributions with a temporal resolution of up to 15 minutes and spatial resolution of about 500m. Best results are achieved for the trace gases NO2, SO2 and O3, due to their good DOAS measurement accuracy. The long term data of the distributions are used to study emission sources in the city, diurnal cycles of the local emission concentrations, chemical transformation processes and the distribution and transport of emission plumes.

The highest trace gas concentrations and lowest spatial variations arose during low wind situations (except for O3 where highest concentrations arise). Emission sources varying in space and time could be distinguished and identified mainly as emissions from traffic (NO2 with O3 depletion) and power plants / industry (SO2). The data directly allow determining the local emission sources. They also show that for the measurement period transport processes to the city were insignificant in comparison to the local emissions. Several insights into chemical processes in the atmosphere could be gained by studying the interrelationship of the measured trace gases. HONO, for example, displayed much lower spatial variability than NO2 and was thus not directly emitted by the same source but rather formed in heterogeneous reactions. It could be shown that transport processes of SO2 and NO2 plumes can be perfectly be studied with this measurement technique. The data were also used to study the spatial variability in daily, weekly and annual cycles of the trace gases.