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High accuracy Primary Reference gas Mixtures for high-impact greenhouse gases

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Climate change, due to increased man-made emissions of greenhouse gases, poses one of the greatest risks to society worldwide. High-impact greenhouse gases (CO_2, CH4 and N2O) and indirect drivers for global warming (e.g. CO) are measured by the global monitoring stations for greenhouse gases, operated and organized by the World Meteorological Organization (WMO). Reference gases for the calibration of analyzers have to meet very challenging low level of measurement uncertainty to comply with the Data Quality Objectives (DQOs) set by the WMO.

Within the framework of the European Metrology Research Programme (EMRP), a project to improve the metrology for high-impact greenhouse gases was granted (HIGHGAS, June 2014-May 2017). As a result of the HIGHGAS project, primary reference gas mixtures in cylinders for ambient levels of CO_2, CH4, N2O and CO in air have been prepared with unprecedented low uncertainties, typically 3-10 times lower than usually previously achieved by the NMIs. To accomplish these low uncertainties in the reference standards, a number of preparation and analysis steps have been studied and improved.

The purity analysis of the parent gases had to be performed with lower detection limits than previously achievable. E.g., to achieve an uncertainty of 2•10-9 mol/mol (absolute) on the amount fraction for N2O, the detection limit for the N2O analysis in the parent gases has to be in the sub nmol/mol domain. Results of an OPO-CRDS analyzer set-up in the 5μm wavelength domain, with a 200•10-12 mol/mol detection limit for N2O, will be presented.

The adsorption effects of greenhouse gas components at cylinder surfaces are critical, and have been studied for different cylinder passivation techniques. Results of a two-year stability study will be presented.

The fit-for-purpose of the reference materials was studied for possible variation on isotopic composition between the reference material and the sample. Measurement results for a suit of CO_2 in air mixtures with varying δ13C values (from -5‰ to -40‰ analyzed with both cavity ringdown spectroscopy (CRDS) and isotope-ratio mass spectrometry (IRMS) will be presented.

Round robins were organized to assess the agreement of the new reference gas mixtures developed by different project partners and to compare the new reference gases with the reference standards currently used by the atmospheric monitoring community (NOAA and AGAGE). These results will also be presented.