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New SI-traceable reference gas mixtures for fluorinated gases at atmospheric concentration

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In order to better support the monitoring of greenhouse gases in the atmosphere, we develop a method to produce reference gas mixtures for fluorinated gases (F-gases, i.e. gases containing fluorine atoms) in a SI-traceable way, meaning that the amount of substance fraction in mole per mole is traceable to SI-units. These research activities are conducted in the framework of the HIGHGAS and AtmoChem-ECV projects.

First, single-component mixtures in synthetic air at ~85 nmol/mol (ppb) are generated for HFC-125 (pentafluoroethane, a widely used HFC) and HFC-1234yf (2,3,3,3-tetrafluoropropene, a car air conditioner fluid of growing importance). These mixtures are first dynamically produced by permeation: a permeator containing the pure substance loses mass linearly over time under a constant gas flow, in the permeation chamber of a magnetic suspension balance, which is regularly calibrated. This primary mixture is then pressurised into Silconert2000-coated stainless steel cylinders by cryo-filling. In a second step these mixtures are dynamically diluted using 2 subsequent dilution steps piloted by mass flow controllers (MFC) and pressure controllers. The assigned mixture concentration is calculated mostly based on the permeator mass loss, on the carrier gas purity and on the MFCs flows. An uncertainty budget is presented, resulting in an expanded uncertainty of 2% for the HFC-125 reference mixture and of 2.5% for the HFC-1234yf mixture (95% confidence interval). The final gas, with near-atmospheric concentration (17.11 pmol/mol for HFC-125, 2.14 pmol/mol for HFC-1234yf) is then measured with Medusa-GC/MS technology against standards calibrated on existing reference scales.

The assigned values of the dynamic standards are in excellent agreement with measurements vs the existing reference scales, SIO-14 from the Scripps Institution of Oceanography for HFC-125 and Empa-2013 for HFC-1234yf. Moreover, the Medusa-GC/MS measurements show the excellent purity of the SI-traceable standards, i.e. the absence of potential contamination from other F-gases to a very low level. These results therefore suggest that this SI-traceable and dynamic method based on permeation and dynamic dilution is a valuable and complementary alternative to the commonly used gravimetric techniques. Finally, we present plans for the future development of a portable generator to allow for an easy on-site calibration with SI-traceable, multi-component reference gas mixtures.