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Atmospheric observations and inverse modelling for quantifying emissions of point-source synthetic greenhouse gases in East Asia

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The fluorinated species carbon tetrafluoride (CF4; PFC-14), nitrogen trifluoride (NF3) and trifluoromethane (CHF3; HFC-23) are potent greenhouse gases with 100-year global warming potentials of 6,630, 16,100 and 12,400, respectively. Unlike the majority of CFC-replacements that are emitted from fugitive and mobile emission sources, these gases are mostly emitted from large single point sources – semiconductor manufacturing facilities (all three), aluminium smelting plants (CF4) and chlorodifluoromethane (HCFC-22) factories (HFC-23). In this work we show that atmospheric measurements can serve as a basis to calculate emissions of these gases and to highlight emission 'hotspots'. We use measurements from one Advanced Global Atmospheric Gases Experiment (AGAGE) long term monitoring sites at Gosan on Jeju Island in the Republic of Korea. This site measures CF4, NF3 and HFC-23 alongside a suite of greenhouse and stratospheric ozone depleting gases every two hours using automated in situ gas-chromatography mass-spectrometry instrumentation. We couple each measurement to an analysis of air history using the regional atmospheric transport model NAME (Numerical Atmospheric dispersion Modelling Environment) driven by 3D meteorology from the Met Office's Unified Model, and use a Bayesian inverse method (InTEM – Inversion Technique for Emission Modelling) to calculate yearly emission changes over seven years between 2008 and 2015. We show that our 'top-down' emission estimates for NF3 and CF4 are significantly larger than 'bottom-up' estimates in the EDGAR emissions inventory (edgar.jrc.ec.europa.eu). For example we calculate South Korean emissions of CF4 in 2010 to be 0.29$\pm$0.04 Gg/yr, which is significantly larger than the Edgar prior emissions of 0.07 Gg/yr. Further, inversions for several separate years indicate that emission hotspots can be found without prior spatial information. At present these gases make a small contribution to global radiative forcing, however, given that the impact of these long-lived gases could rise significantly and that point sources of such gases can be mitigated, atmospheric monitoring could be an important tool for aiding emissions reduction policy.