



Halogenated greenhouse gas emissions over Central Europe inferred from ambient air measurements and 222-Rn activity

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To check for compliance with the reduction targets defined under the Kyoto Protocol, each participating country has to report its greenhouse gas emissions to the UNFCCC (United Nations Framework Convention on Climate Change). These emissions are calculated using a bottom-up approach, by combining categories of compound use and specific activity (release) functions. The uncertainties of these estimates are not well defined, thereby making an independent validation of the reported emissions highly desirable. Top-down estimates based on atmospheric concentration measurements and using a reference species as a priori information are a promising method for independent emission estimates. For this purpose, atmospheric Radon (^{222}Rn) is very well suited due to its exactly known radioactive decay lifetime of 5.5 days and its homogeneous release over soil with comparatively small spatial and temporal variability.

In the present study, concentration measurements of halogenated greenhouse gases such as hydrofluorocarbons (HFC), perfluorocarbons (PFC) and SF₆ at the remote sites Jungfrauoch (Switzerland) and Mace Head (Ireland) were combined with backward calculations of the Lagrangian Particle dispersion model FLEXPART to derive emission rates over Central Europe. The ability of FLEXPART to simulate the origin of air masses arriving at the receptor point was checked using ^{222}Rn measurements in combination with the flux map recently developed by Szegvary et al. (2007), and analysis was restricted to episodes where FLEXPART successfully reproduced the observed concentration pattern of ^{222}Rn . This procedure not only removes all measurements where the flow regime of air masses is uncertain and source attribution of emissions is therefore difficult, but also allows to correct for potential model uncertainties originating e.g. from the complex topography not resolved by the model.

The top-down estimations derived in this study generally agree well with the bottom-up estimates submitted to the UNFCCC. Exceptions are HFC-23 (byproduct of HCFC-22 production) and HFC-152a (foaming agent), where Radon-derived emissions are much higher than the bottom-up estimates. This result suggests that the bottom-up approach might be underestimating or missing some significant sources of greenhouse gas emissions.

References

Szegvary T., Leuenberger M.C., Conen F. Predicting terrestrial ^{222}Rn flux using gamma dose rate as a proxy. *Atmos. Chem. Phys.* 7 (11), 2789–2795, 2007.