# EVALUATION OF CO AND $SF_6$ AS QUANTITATIVE TRACERS FOR FOSSIL FUEL $CO_2$: THE MODELLERS' VIEW

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## ABSTRACT
Simulations with a regional transport model are evaluated in order to determine to which extend the indirect fossil fuel combustion tracer CO or the purely anthropogenic tracer $SF_6$ can be used to retrieve the contribution of fossil fuel emissions in the atmospheric $CO_2$ signal.

## INTRODUCTION
The separation of the contribution from fossil fuel emissions in the atmospheric $CO_2$ signal is crucial for the assessment of continental carbon fluxes from inversions of atmospheric $CO_2$ concentration measurements. Though fossil fuel emissions are mapped by inventories, they are not yet verified by independent measurements. The radioactive carbon isotope $^{14}C$ in $CO_2$ has proven to be a good quantitative tracer for the fossil fuel component in atmospheric $CO_2$ because fossil fuels do not contain any $^{14}C$ [*Levin et al., 2003*]. But the analysis of $^{14}CO_2$ is complex and does not allow continuous observation with high temporal (i.e. hourly) resolution. Carbon monoxide is another potential tracer for fossil fuel $CO_2$ because it is emitted concurrently with $CO_2$ during combustion processes. CO has the advantage that continuous measurements are comparatively cheap and easy. However, CO has atmospheric sinks and also sources different from fossil fuel burning and the CO/fossil $CO_2$ emission ratio varies in space and time depending on the mixture of source types. Another surrogate tracer is the long-lived $SF_6$, which is closely linked to human activities but not explicitly to fossil fuel emissions.

## MODEL SETUP AND EMISSIONS INVENTORIES
The regional atmospheric transport model REMO [*Chevillard et al., 2002*] is used to simulate the temporal and spatial distribution of fossil fuel $CO_2$, CO and $SF_6$ mixing ratios. In the current set up the horizontal grid resolution is 55 km x 55 km and the model domain covers a large part of the Northern Hemisphere (north of 30ºN). To account for contributions from sources outside the model domain REMO is nested into the global transport model TM3 [*Heimann and Körner, 2003*]. Additional to surface fluxes, which are prescribed from inventories and biosphere models, also the photochemical processes for CO in the atmosphere are included in REMO. CO and $CO_2$ emissions from fossil fuel combustion were extracted from two different emissions inventories: (1) The Emission Database for Global Atmospheric Research (EDGAR), which provides annual mean emissions for several base years on a global 1°x1° grid [*Olivier and Berdowski, 2001*], and (2) hourly emission values calculated by the Institute of Energy Economics and Rational Use of Energy (IER) on a 50km x 50km grid for the greater part of Europe available for the year 2000 [IER, emission data, 21 Dec. 2004]. A comparison of annual mean CO and $CO_2$ emissions and resulting emission ratios in Europe reveal large regional differences between the two datasets because estimates are based on different data sources and also the spatial pattern of emissions and hence emission ratios is strongly dependent on the way national totals are disaggregated. $SF_6$ emissions were also extracted from the EDGAR database.

## COMPARISON WITH OBSERVATIONS
REMO simulation results are first compared to continuous measurements of CO, $CO_2$ and other trace gases at several stations in Europe in order to investigate model performance. As an example, observed $CO_2$, CO and $^{222}$Radon time series for August 2002 in Heidelberg are shown in Fig. 1 together with REMO results from simulations using the two different emissions inventories for fossil fuels. In summer the model often overestimates the mixing ratios of all three gases, in particular during nighttime. While both model simulations are quite similar for $CO_2$, EDGAR emissions result in CO mixing ratios almost twice as high as IER emissions. Since the mean radon source prescribed in the model (52 Bq $m^{-2}$ $h^{-1}$) is only slightly lower than observed long-term mean emissions in the Heidelberg area, the overestimation indicates a tendency of the model to underestimate vertical exchange during night in this grid cell. But even after accounting for this systematic effect it still seems that both inventories, in particular EDGAR, overestimate CO (and $CO_2$) emissions in summer.

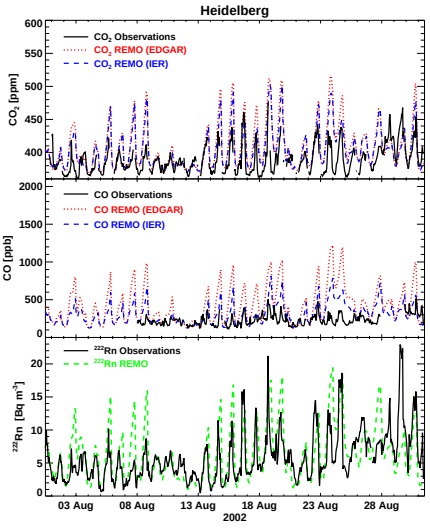


Fig 1. Hourly $CO_2$ and CO mixing ratios and $^{222}$Rn activity in Heidelberg in August 2002, comparison of REMO results with observations.

At the Heidelberg site quasi-continuous nighttime observations of $^{14}CO_2$ are available, which allow to determine the fossil fuel $CO_2$ contribution at a two-weeks temporal resolution (compare accompanying paper by Levin et al., this issue). Respective values have been extracted from REMO simulations for the same time periods in 2002 as for the integrated observations (Fig.2a). In order to reduce model transport uncertainties and hence allow a more quantitative evaluation of emissions inventories, CO as well as fossil fuel $CO_2$ from both simulations were normalized using the $^{222}$Radon observations. The overestimation of CO by both model simulations is still evident (Fig.2b) but also fossil fuel $CO_2$ is considerably higher than in the observations in summer and early autumn (Fig.2a). The atmospheric $CO/CO_2$fos ratio also shows a systematic difference between the two simulations (Fig. 2c). Most probably the emissions inventories postulate too high CO as well as too high $CO_2$ emissions, which could either be due to an overestimate of sources or emissions factors or both. If the calculated mixing ratio is normalized with the measured CO, simulated $CO_2$fos compares much better with observations (Fig. 2d), indicating a way to correct inventory-based model simulated fossil fuel $CO_2$ contributions at sites where CO but no $^{14}CO_2$ observations exist. Based on our current simulations this would result in mean relative errors of 20% and 40% for EDGAR and IER emissions, respectively.

## CONCLUSIONS

Model simulations provide a useful tool to investigate possibilities of temporal and spatial propagation of a $^{14}$C calibration of CO- (or $SF_6$-) derived fossil fuel $CO_2$ at selected stations. However, the inter-comparison of simulated results based on different emission inventories and also the comparison with observed $CO/CO_2$fos ratios reveal the large uncertainties still existing in the available emission inventories.

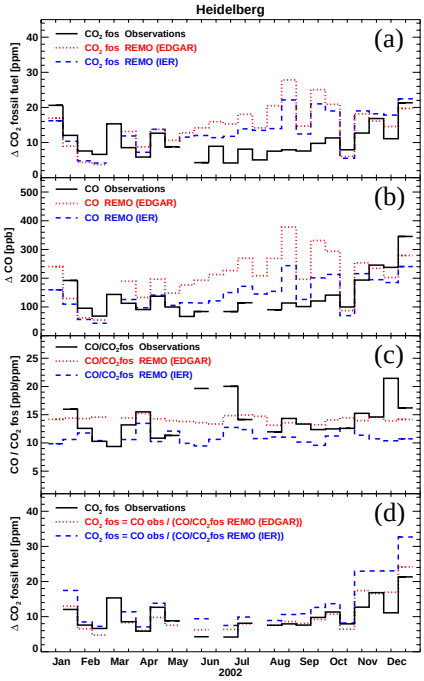


Fig. 2: (a) Fossil fuel $CO_2$, (b) CO, both normalized using $^{222}$Rn, (c) $CO/CO_2$fos ratios and (d) fossil fuel $CO_2$ recomputed from observed CO and simulated $CO/CO_2$fos for two-weekly integrated samples in Heidelberg, comparison of REMO results with observations.

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