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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Emissions of methane in Europe inferred by total column measurements" *by* Debra Wunch et al.

Anonymous Referee #1

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The paper by Wunch et al. attempts to estimate methane (CH_4) emissions in Europe from a network of five ground-based sun-viewing total column spectrometers operated within the TCCON (Total Column Observing Network). The approach builds on calculating pair-wise differences between the (detrended) CH_4 and carbon monoxide (CO) concentrations at the five observatories and assuming that CH_4/CO emissions ratios scale like the detected concentration anomalies and that the input CO inventory is accurate. The authors conclude that two state-of-the-art CH_4 emissions inventories overestimate emissions in Central Europe.

I have major concerns detailed below which make me doubt that the results are robust and publishable in ACP. If I misunderstood things, please consider addressing these

points in greater depth by the (currently quite short) manuscript:

1. By design of the method, the CH_4 emissions estimate cannot be more accurate than the CO inventory used for scaling. A priori, I would argue that CH_4 inventories are per se better than CO inventories since there is less variability in CH_4 sources and less changes in source patterns. Figure 3 also indicates that the EDGAR and the TNO inventories show much larger discrepancies for CO than for CH_4 (likewise text p.6, l.21 ff). Then, the authors need to extrapolate the CO inventories under changing background conditions (change of source type) inducing further uncertainties. What is the total CO-induced error on your CH_4 emission estimates? Are the conclusions robust against this error? The conclusion needs to be quantitative in that respect.

2. The CH_4 and CO concentration anomalies are calculated pair-wise among the five stations. The ratio of the concentration anomalies is then used to scale the CO emission inventories to yield CH_4 emissions estimates. This requires that the concentration anomalies are representative of emissions inbetween the stations. In my understanding this requires that CH_4 and CO are emitted (roughly) coincident in time and space and that they share transport between stations. What is the temporal correlation of CH_4 and CO sources? What is the spatial overlap of CH_4 and CO sources (quantitatively)? Does CO (e.g. in the urban boundary layer) survive sufficiently long such that loss of CO over, say, 5-10 days is entirely negligible?

3. The analysis is based on five stations in Central Europe. As stated above, the analysis requires that the pair-wise anomalies are representative of emissions inbetween the stations. In my understanding, this implies that airmasses passing an upwind station should (at least for a large fraction of days) pass a downwind station. Is the network sufficiently dense to avoid a significant number of upwind airmasses being never sampled by a downwind station?

In fact, the time-lag analysis (Fig. A2, calulcating pair-wise anomalies not for the same day but for a time lag of several days) does not show any distinct dependence on time lag. This means that there is no indication that airmasses passing upwind stations also pass downwind stations (more often than sporadically).
Likewise for being representative of the entire Central European area (incircled by the stations), the five stations would need to sample (on a frequent basis) air masses that actually sampled the entire area. Is this condition met?
Is there evidence that these sampling issues are not significant?

The above concerns a touched on by the manuscript but I would argue that they are so critical that they require quantitative answers not just handwaving arguments. In general, my concerns 2 and 3 could be addressed by a comprehensive simulation study based on CH_4 and CO emissions inventories, a chemical transport model and an observation operator that mimicks the sampling by the ground-based spectrometers. I would recommend implementing such an approach to invalidate my concerns and to verifiy the assumptions or to reconsider the methodology.

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