Atmos. Chem. Phys. Discuss., 8, S97–S101, 2008
www.atmos-chem-phys-discuss.net/8/S97/2008/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Ten years of CO_2, CH_4, CO and N_2O fluxes over Western Europe inferred from atmospheric measurements at Mace Head, Ireland" *by* C. Messager et al.

A. Hirsch (Referee)

adam.hirsch@noaa.gov

Received and published: 6 February 2008

General comments:

This paper is well within the scope of ACP. It is well written and interesting. While the radon tracer technique they use is not new, it is applied in a new way, namely the long term monitoring of European greenhouse gas emission trends. The authors argue that while there may be systematic biases in the technique, these biases are stable over time, allowing trends to be discerned.

By using the central limit theorem and focusing on annual average fluxes, they are able to reduce the uncertainty in their derived fluxes to levels that allow trend detection over the course of a decade (see specific comment 1, below, though). They conclude that inventory estimates of reductions in methane, carbon dioxide, and nitrous oxide fluxes from 1996-2005 are supported by their data. The paper is well set in the context of past research, with the results compared to both inverse modeling estimates and inventory estimates. Their careful identification of European or UK/Ireland "events" using trace gas and meteorological data is verified using a Lagrangian particle dispersion model. Their fluxes also agree well with independent inverse estimates using the same model (NAME). When the radon tracer technique does not agree with the inventories in absolute amount, this disagreement is used to their advantage, such as quantifying the magnitude of natural methane sources, which are not included in the inventory estimates. They even use their diagnosed carbon monoxide fluxes and an inventory estimate of CO/CO2 emission ratio to divide their total carbon dioxide source into fossil fuel burning and natural components, the former agreeing fairly well with the inventories and the latter agreeing fairly well with a biogeochemical model estimate of respiration. As the authors point out, the agreement of the radon tracer method and the inventories in the case of nitrous oxide is puzzling, signifying either that anthropogenic emissions are overstated or natural emissions are minimal. The authors also address the shortcomings of their technique in detail. They map out a path to address some of these limitations, such as using back trajectories to better identify the geographic origin of the signals measured at Mace Head.

The beauty of the radon tracer method is that it does not rely on a chemical transport model to calculate flux estimates. It simply requires a knowledge of the radon flux and good correlation between radon and another trace gas. In general, the uncertainty of the radon flux is thought to be a shortcoming of the technique. Improved estimates of the radon flux across space and time will help diagnose whether the assumptions made here regarding the radon flux are reasonable. As long as there are enough events sampling the continent somewhat randomly, the variability of the radon flux is less of a concern (due to the central limit theorem), unless there is really some long term pan-European secular trend in radon flux due to, say, longterm changes in soil moisture. The underlying production of radon is not going to change. There are concerns here about temporal and spatial representivity. If fluxes have a seasonal cycle and the majority of "events" occur in the fall and winter as stated on page 1208, the annual average will obviously be weighted toward fall/winter values. This issue is especially problematic for carbon dioxide, with its large seasonal cycle. It is possible that trends in fluxes are therefore more representative of trends in winter fluxes rather than annual fluxes. Likewise, if there are large spatial differences in fluxes, and the majority of events originate in one part of the continent (as happened in 1999-2002), the "European" flux will be weighted to the value in those countries. It is not clear to what degree the 1999-2002 increased contribution from Germany and Netherlands affects the trends presented here. My sense is that the authors are just taking the difference between the start and end of their record and are not so concerned with the actual trend between the two. Could you comment on this?

Specific comments:

The major conclusion of the paper is that "the decrease of the anthropogenic CH4, CO and N2O emissions in Europe estimated by inventories...is confirmed by the Mace Head data within 2%." I think this might be a bit of an over-statement of the precision of the radon-tracer method. Please put some error bars on the flux decreases and a level of significance on the decreases. Also, please describe how you calculate the decrease - is it a linear trend or just 2005 minus 1996? I would think the interannual variability would make it difficult to get a linear trend from 1996-2005 with a significant slope. Actually it looks like in general European fluxes rise or are steady from 1996-2000 and then decrease quickly from 2000-2004, while the inventories continually decrease. Would you say that you disagree with the inventories during the first part of the record?

Also, an agreement between your technique and the inventories is interpreted in your paper as meaning that your are capturing changes in anthropogenic fluxes and that there is no trend in natural fluxes. Do you think it is possible that there have been trends in natural fluxes? This is claimed for CH4 in Ireland/UK - why shouldn't it be possible

ACPD

8, S97–S101, 2008

Interactive Comment

for the Continent, too? The paper would be strengthened if there were process based model estimates of CH_4 and N_2O fluxes to address this. As an aside, it might be helpful to readers to describe which N_2O fluxes are natural versus anthropogenic (e.g. agricultural fertilizer use, animal waste).

page 1194 - what do you mean by "scaled" in "The fluxes scaled over Europe..."?

page 1195 - do you mean the "relative" error instead of the "absolute" error?

I think the discussion of the synoptic meteorology that brings local Irish versus background versus European air is very good, especially in combination with figures 5 and 6.

page 1199 - it is unclear to me what the HYSPLIT-4 analysis adds in addition to the very extensive NAME analysis. Please clarify.

page 1200 - what do you think causes the small diurnal cycle in CO_2 for the European event - are you seeing the diurnal cycle over the continent transported to Mace Head or is there some influence from England/Ireland soon before the measurement?

page 1202 - a minor point - you use the time for the air to get back to the continent to correct for radon decay. That gives you the original radon concentration before it left Europe. However, in the radon tracer method do you also need to consider the radon decay that occurred during its transport over the continent? In other words, does the radon tracer method require that radon be converted into a conserved tracer? Hirsch et al. 2006 showed there could be a 10-20% difference if radon were modeled with or without radioactive decay. This is just something you might consider at some point - I don't think you need to address it here.

page 1205 - does the Bousquet [2006] paper actually claim that wetland emissions increased by 35% in Ireland and the UK?

page 1212 - are you meaning to say the covariance between radon and other species emissions or covariance between other species emissions and surface influence?

ACPD

8, S97–S101, 2008

References - several papers (Szvegvary, Hirsch) have final ACP versions. Two Szegvary papers are referenced in the paper, only one is included in the references.

Table 5 - Should be CH_4, not CO_4.

Figures - I didn't find that Figure 2 added very much to the discussion. Same with figure 4. Why do you think the diurnal cycle in the lower right hand figure of figure 3 is so different for NAME versus your selection criteria?

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 1191, 2008.

ACPD

8, S97–S101, 2008

Interactive Comment