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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.

Anonymous Referee #1

Received and published: 10 February 2008

This paper presents a 10-year "climatology" of CO_2, CH_4, CO and N_2O surface fluxes for western Europe and Ireland+U.K., derived from the ratios between the concentrations of these substances and corresponding radon concentrations. The method is independent of model assumptions (except for some classification of the data) and based exclusively on measured quantities. As a disadvantage it has limited (and poorly defined) spatial resolution and depends on the assumption that radon fluxes are known and spatially and temporally relatively invariant. The paper is generally well written and worthy of publication in ACP. However, I have a few concerns as detailed below, which should be addressed before the paper can be published in ACP. Furthermore, I ask the co-authors who are native English speakers for a thorough elimination of flaws as-

sociated with the English language before the submission of a revised version of the paper.

My major comments are: Title: write "surface fluxes" instead of "fluxes" as "fluxes" alone is not specific enough. It could also mean, for instance, meridional fluxes.

My major concern is with the assumption of constant and uniform radon fluxes. To what extent does this assumption supported by measurement data and what errors do occur as a result from that assumption. Could a formal error propagation be done?

Another major concern is that slopes between concentrations of greenhouse gases and radon concentrations have been determined from individual events. Events have been eliminated when correlations have been low. What happens when during one event the source region changes, e.g., when air masses first originate from a high-emission area and later from a low-emission area, with vastly different greenhouse gas emissions but similar radon emissions (if the assumption above is correct). This would cause a low correlation for such an event, thus rendering the individual slope rather meaningless and leading to the elimination of such cases. In my opinion, such events would be rather typical, as the area of influence would often sweep over Europe just before or after synoptic patterns change. But what is the justification for removing them from the data set?

What is the uncertainty of the derived trends? The trends appear to be non-uniform with, e.g., N_2O emission increases from 1996-2000, followed by emission decreases. This is not seen in the UNFCC data and I doubt that the resulting overall decrease over the entire period, which happens to be close to the UNFCC total decrease, is significant. Please clarify!

On page 1203, lines 4-6, the authors say that the uncertainty of the radon exhalation rate is 20

When Europe is covered by snow in winter, radon fluxes would cease. Could, thus,

ACPD

8, S147–S150, 2008

Interactive Comment

snow-cover information be used for improving the method?

The data set has been split into marine, European and Ireland+UK influences. I am wondering to what extent North American influences can be seen in the marine data set and how this would affect the interpretation of marine as a baseline.

Local wind speeds have been used as a criterion to distinguish between air masses of different origin. However, the local wind speed is not necessarily representative of the transport from, e.g., western Europe, since the wind field is instationary and inhomogeneous and, furthermore, long-range transport may occur also above the PBL. I think it would have been better to use wind data (e.g., from ECMWF) that have been averaged over some upwind fetch and/or time period before the measurement to better characterize the flow prior to arrival.

The authors point out that land breezes are important for transport of trace substances from Ireland to Mace Head. I am not convinced that land breezes are so important at this rather high-latitude marine site where synoptic-scale winds should dominate most of the time. Is there a reference to support your statement?

For Fig. 5+6, I suggest that color is used. The various black and grey lines and grey shadings are difficult to read.

The sequence of tables 4-11 should be in the same order as they are discussed, i.e., as the sections 6.1-6.4 (methane first,etc.)

Page 1209, line 8: I don't think the statement that there are no natural CO sources is true. See Forster et al. (2001) for influence of boreal forest fire emissions at Mace Head, and Witham (2007) for influence from fires in eastern Europe.

Forster, C. et al. (2001): Transport of boreal forest fire emissions from Canada to Europe. J. Geophys. Res. 106, 22,887-22,906.

Witham, C., Manning, A. (2007): Impacts of Russian biomass burning on UK air quality. Atmos. Environ. 41, 8075-8090.

ACPD

8, S147–S150, 2008

Minor points, language: In abstract, line 12 and similar occurrences at many other places throughout the entire manuscript: 4.1±1.5 10^6 should be 4.1±1.5×10^6

Page 1193, line 2: mixing ratioS

Singular and plural has been mixed randomly throughout the manuscript. Example: page 1194, line 2: "All this studies" should be "All these studies"

Page 1198, line 2: absolute difference: do you mean absolute mean difference?

Page 1198, lines 3-4: I do not understand the text in the bracket. A value cannot represent a certain wind speed (unit m/s) and a frequency (no unit) at the same time.

Page 1200, line 1: irish -> Irish, european ->European

Caption to section 5: Fluxes estimation method -> Flux estimation method

Page 1201, line 17-18: citations should be taken out of brackets

Page 1203, line 10: Table -> TableS

Page 1203, line 23: countries8217; -> country's

Page 1204, line 2: eliminate the word "degree" as you have already used degree symbols

Page 1205, sentence starting at line 20: We compared8230; this sentence is incomplete; same for last sentence of section 6.1

Page 1210, line 3: ratio CO/CO2 ratio: eliminate one "ratio"

Tables 5 and 9: CO_4 -> CH_4

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