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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Abundance and Sources of Atmospheric Halocarbons in the Eastern Mediterranean" *by* Fabian Schoenenberger et al.

Anonymous Referee #2

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Abundance and Sources of Atmospheric Halocarbons in the East Mediterranean

Fabian Schoenenberger et al https://doi.org/10.5194/acp-2017-451

This manuscript describes a study using high-frequency, in situ measurements of halocarbons (HCFC-22, HCFC-142b, HFC-134a, HFC-125, HFC-152a, and HFC-143a) at four sites in Europe, along with an atmospheric inversion model, to estimate regional emissions in 2013. The authors use measurements made over 6 months at Finokalia on the island of Crete (Greece), in conjunction with three other sites in Europe to obtain information on emissions sources in the Eastern Mediterranean, typically not well-captured by measurements at the other three sites (Mace Head, Ireland (MHD); Jungfraujoch, Switzerland (JFJ); Monte Cimone, Italy (CMN)). The paper describes various model sensitivity runs in which model parameters (covariance, uncertainty of

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prior emissions) were varied. The authors also explored how the addition of the Finokalia measurements impacts inversion results compared to runs using data from MHD, JFJ, and CMN).

This paper makes an important contribution to previous work estimating European emissions of halogenated greenhouse gases and Montreal Protocol gases using atmospheric observations (topâĂŤdown method). Measurements at Finokalia provide additional constraint and improve emissions estimates from countries in the Eastern Mediterranean, including Greece and Turkey and portions of North Africa.

General Comments:

This paper is well-written and results are well-presented.

While I don't have any problem with the methods, as the inversion technique is well-established and has been used previously to interpret similar data from MHD, JFJ, and CMN, I do have some comments regarding the results.

1) Since your period of observation is rather short (6 months), you are unable to provide any information about the seasonality of emissions. I suggest you include the potential for seasonality in your discussion. Others (Hu et al 2017; Grazioisi et al 2015, Xiang et al, 2014) have suggested that emissions of some HCFCs and HFCs show seasonal variations, with higher emissions in summer compared to winter. That, coupled with the fact that the Finokalia source sensitivity region was different between winter/spring and summer (your Figure 2), suggests that you could be missing a component of the mean annual emissions (or even the mean for 6 months) because you are less sensitive to emissions from some areas in summer, when higher emissions are more likely. This might be particularly true for Egypt. 2) You ran sensitivity studies in which the uncertainties associated with prior emissions were varied, and other sensitivity studies that explored different covariance treatments. However, you did not test the sensitivity of the magnitude of the prior emissions or distribution of priors. I think you should either include model runs with higher and lower priors, or provide some justification for why

this is not needed. Is simply changing the covariance treatment and prior uncertainty enough? 3) Can you comment on the sensitivity to background assignment? Is the choice of background more or less important compared to treatment of covariance? Did you try different background methods other than REBS? 4) Minor: It would be helpful to see (in the Supplemental) a time series similar to figure 3, but expanded over a few days or weeks. This would show more clearly the duration of "pollution" events at the different sites and provide a qualitative picture of the "signal". This would also provide some information about correlations among different halocarbon species. For example, you see what looks like pollution events for HFC-143a at Finokalia in December, but do not see corresponding pollution events for HFC-125, even though some refrigerants, such as R404A, are a blend of both compounds.

Specific Comments:

Line 50-51: CH3Cl is probably an exception to the statement that all long-lived halocarbons are potent greenhouse gases (CH3Cl lifetime is $\sim$1 yr, but 100-yr GWP is only 11). Consider chaging "all" to "most" or "many".

Line 71: prefer "base value" to "original value"

Line 139: Is the air handling system (pump, drier) also similar to what is used at CMN? It would be helpful to specify here.

Line 202: Why not release particles at 3400 or 3500 m, closer to the actual site JFJ elevation? Is this not possible?

Line 301: Consider: "We followed three different strategies concerning the design of covariance matrices . . .. . ."

Line 429: I find it hard to see "satisfactory performance of the transport model" in figure 4. You might rephrase in terms of comparison to other studies, i.e. is this level of reproduction of variability typical for FLEXPART?

Line 467: remove comma in "driven by, an increase"

Line 495: remove comma in “which shows, that”

Line 472: Update Brunner et al 2016 to Brunner el al 2017 as this paper is now available.

Line 573: Do you mean “their mean values OR the analytical a posteriori. . ..”?

Line 602: remove comma in “regions, defined by”

Line 619: maybe a comment on domain emissions compared to global total (Simmonds et al 2017) Section 3.4.2: You might consider comparing Brunner et al 2017 HFC-125 estimates for Italy (1.2 Gg with your 1.05 Gg estimate)

Line 680: comma not needed in “fact, that”

Table 3 Caption: Line 1102: “ in in Mg yr-1” Duplicate word, and I think you mean “Gg yr-1”

Figure 8 caption: Is “average mean” redundant? Are uncertainties 2-sigma here also?

Figure 9 caption: Better to cite Harris and Wuebbles (2014), as GWP were calculated in Chapter 5 of the 2014 Ozone Assessment, rather than in Chapter 1, Carpenter and Reimann

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