Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-451-RC1, 2017 © Author(s) 2017. This work is distributed under the Creative Commons Attribution 3.0 License.



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

## **Anonymous Referee #1**

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General comments: The authors reported measurements of HFCs and HCFCs at a new site located in Finokalia in the Eastern Mediterranean and compared with measurements at three AGAGE sites in the western and central Europe. The authors used these measurements and estimated HFC and HCFC emissions from the Eastern Mediterranean and Europe using inverse modeling. It is nice to see that the authors considered different inversion configurations (i.e., different prior error covariances, different prior uncertainties and different observations) and tested the sensitivity of derived emissions to these different configurations. However, there are a few important tests the authors did not perform to fully demonstrate the robustness of their derived emissions: (1) the prior emissions the authors use are always biased high. I have concerns whether these biased priors may result in posterior emission estimates that

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are also biased high, especially for an inversion configuration that do not consider correlated errors in the prior emissions (e.g., S-MS). The authors may consider including priors that are biased low and see if they obtain similar posterior emissions. (2) The authors derive temporally constant emissions from their inversion framework. This may result in larger model-data mismatch errors and an under-fit of their atmospheric data comparing to inversions that estimate time-varying emissions. This problem could be more severe for chemicals that have strong seasonal cycles in their emissions (e.g., HFC-134a, HCFC-22, and HFC-125). Conducting an inversion with deriving varying emissions could be beneficial to further test the robustness of their results.

## Specific comments:

Lines 25-27 "The eastern Mediterranean is home to ...under the Kyoto and Montreal Protocol". This sentence seems unnecessary in the abstract.

Lines 42-44 "reduction of the uncertainties by 40-80%". This reduction is for which region?

Line 51 "all long-lived halocarbons are potent greenhouse gases". This is inaccurate. Gases such as CH3Br and CH3Cl are not potent greenhouse gases.

Lines 57-59 "To track the development of CFC and HCFC emissions to the atmosphere,..." I don't think the reporting required by the MP is to specifically track emissions.

Line 71 "15% of the original value". Inaccurate. Maybe consider mentioning 15% of the baseline value during the years of xx?

Line 168. Please spell out "NNE-E"

Line 183 "three-hourly aggregates". Do you mean "three-hourly averages"?

Lines 200 - 207 Transport simulation. Why do the authors choose to run their transport simulation at altitudes that are different from their actual sampling altitudes? How

much different are the derived sensitivities at their simulation altitudes and the actual sampling altitudes?

Lines 210 - 212: 1.7% degradation of HFC-152a emissions. Is this for the summer or an average for the whole year? Please be clear.

Lines 289 – 290. HCFC-142b is mostly used as a foam blowing agent whereas HCFC-22 is used as refrigerant in air conditioning. So, they are not necessarily collocated. Assuming they have the same regional emission shares is not a good assumption for creating HCFC-142b prior emissions.

Line 293. Which version of the EDGAR inventory do you mean? Be clear.

Lines 307 – 321. Uncertainty covariance matrix B. Please explain why giving correlation lengths of 200 km and 5 days? How sensitive are the derived emissions to these correlation lengths? Also be clear on whether you considered anti-correlated errors between BE and BB.

Lines 325 – 327. Calculation of  $\sigma$  model was derived based on the prior simulation and observations. This approach would likely overestimate the model-data mismatch errors and result in an under-fit of atmospheric data.

Lines 368 - 375. Please indicate in the paragraph whether you consider results from S-NFKL and S-OFKL into the final emission uncertainty estimation.

Lines 403 – 404. Inaccurate description on HFC-134a and HFC-125 usage. Only HFC-134a is mainly used in mobile air conditioning. HFC-125 is mainly used in commercial refrigeration and residential air conditioning.

Lines 416 - 418. It seems that the baseline was shifted at FKL for HCFC-142b in Fig. 3. What caused it?

Lines 428 - 429. "simulated a priori mole fractions reproduced the variability of the observations". Please provide the correlation (r2) value between the simulated and

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observed mole fractions here. In this way, it gives quantitative information on how much variability the prior simulation explained the observed variability.

Lines 495 – 497. Error reductions were expected after an inversion. This cannot demonstrate that you achieved satisfactory emission estimates.

Lines 510 - 520. The authors discussed the differences in RMSE, R2 and Taylor Skill Scores for inversion results obtained from the "global" and "local" approaches. But how different are the covariance parameters derived from both approaches? It would be useful to show and discuss those differences first.

Lines 521-529. The results show the S-MS setup improves RMSE, R2 and Taylor Scores. This is, to some extent, expected and pre-defined, because the S-MS scenario did not include correlated errors in the R matrix whereas the other two scenarios (base and S-ML) did. To really test and understand the advantages or disadvantages of having correlated errors in the prior fluxes or atmospheric data, it is better to isolate the problem. In another word, it is better to have a scenario that has a same R matrix as the S-MS scenario and a same B matrix as the base scenario.

Lines 530 - 540. Results seem to indicate emissions derived from the S-MS may be more biased toward the prior.

Lines 605-607. It is a little dangerous to conclude a trend from two different studies given unknown differences from different methodologies and different atmospheric data.

Line 628. "account for 39.7%". Should this be  $\sim$ 50% (0.53 / 1.0)?

Lines 628 - 632. "... a reduction by a factor of 2". Again, it is not a good idea to conclude a quantitative trend from two different studies.

Lines 644 – 646. Inaccurate description on the HFC usage. HFC-125 is often used with HFC-143a in commercial refrigeration or with HFC-32 as a refrigerant blend in residential air conditioning.

Lines 666 - 668. Citation of Brunner et al. [2016] here and thereafter. It is better not to cite an article that is not publicly available.

Line 695. HFC-143a is mainly used in commercial refrigeration, not in air conditioners.

Lines 717 - 719. The incomplete reporting to the UNFCCC does not seem to be an appropriate explanation for much larger reported emissions than atmosphere-derived emissions.

Lines 727 - 733. The authors compared their emission estimates with a few previous top-down emission estimates. Although the authors noted the differences, they did not provide an explanation why they are different.

Lines 806 - 808. Why including FKL increased the estimation of the baseline mixing ratios at JFJ and CMN?

Please also note the supplement to this comment: https://www.atmos-chem-phys-discuss.net/acp-2017-451/acp-2017-451-RC1-supplement.pdf

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-451, 2017.