Review AMT-2018-21: IASI nitrous oxide……by Y. Kangah et al

The paper deals with an application of IASI data to the retrieval of N_2O. The authors analyzed IASI data for the Northern-Hemisphere summer season (June-July) in 2011 and claim that the relatively high concentration of N_2O over the Eastern Mediterranean basin is a result of pollution transport from Asia. I think that the paper shows potentially arguing and interesting results. However, an in-depth and accurate reading of the document shows, and I am sorry, that the paper presents a lot of weak points and many technical aspects, which need to be clarified and properly addressed before this study can be accepted for publication.

General Comment

It is known that the Mediterranean summer (June to September) is characterized by high pressure over the Mediterranean Europe and a low-pressure trough extending from the Persian Gulf through Iraq to the southeastern Mediterranean (see e.g., Y. Goldreich, Springer, 2003). It is now very well understood (e.g., Karnieli et al. JGR, 2009 and references therein) that this kind of weather pattern yields persistent northwesterly winds which causes long-range transport of air masses and pollutants from southeastern and southwestern Europe into the eastern Mediterranean basin. In agreement with this weather pattern, previous IASI studies (also cited in the present paper, e.g., doi:10.1364/OE.21.024753) have indeed evidenced higher concentrations of green-house gases over the Eastern Mediterranean basin. Conversely, the authors suggest that there could be another atmospheric pathway along which pollutants are transported to the Eastern Mediterranean basin. Because of the importance of this finding, the authors should be much more convincing in showing that their methodology has no weak points. In effect, their analysis is based on N_2O profiles retrieved with less than 1 degree of freedom, and they concentrate on N_2O layer average at ≈309 hPa, but they fail to show that this layer average has been independently resolved of the rest of the profile. In view of the broad structure of N_2O AK they provide in the study, it is likely that they are mostly sensitive to the column amount of N_2O.

I have detailed my points below, which I hope can help authors to improve the paper.

Major remarks

1. Page 4, line 17. NEDT depends on the scene temperature, which, because of atmospheric absorption, is wave number dependent. Was this dependence taken into account? By the way, I suggest that the comparison should be made in the radiance space, using NEDN, which is wave-number independent.

2. Page 5, Equation (1). This equation should be introduced this way…*We used optimal estimation based on the Levenberg-Marquardt (put reference) algorithm as modified by Fletcher (put reference) and adapted for Optimal Estimation by Rodgers.* By the way, the important aspect here is that Equation (1), as it is written, is wrong. The term multiplying the leftmost S_a^{-1} should be $(1+\gamma)$ not simply, γ. In fact, Eq (1) should transform back to the OE estimator for $\gamma = 0$, which is not the case. Hope this is just a typo. Furthermore, how γ is chosen at each step? Do authors perform retrieval in the BT space or radiance space? Please, clarify.

3. Page 5, Equation (2) is not consistent with Eq. (1). Apparently, the authors use $\gamma = 1$ for the final iterate, but then Equation (1) is not the correct OE estimator, and the final iterate would depart from optimality.

4. Page 5, Equation (3) applies just to one parameter. Considering that the authors claim to use a simultaneous approach, how is the a priori covariance of the whole state vector built up?

5. Page 5, line 20. Please show the N_2O profile. The retrieval approach is strongly depending on such a background.

6. Page six, line 2. As before, show the CO_2 profile and those of other species used as a priori reference.

7. Page 6, line 13, what is a sink parameter? Please, explain.

8. Page 6, Equation (4). The denominator is wrong. The degrees of freedom of the χ^2 form are dim(*Y*). This can be demonstrated by a trivial use of the Standard Theorem of Least Squares (e.g., Rao 1973, the authors should consider that $\hat{X}$ is estimated from the data, so that the remaining degrees of freedom of data are dim(*Y*)- dim($\hat{X}$) and dim($\hat{X}$) + [dim(*Y*)- dim($\hat{X}$)] = dim(Y)). Since the authors use a retrieval algorithm for which dim($\hat{X}$)≈dim(*Y*), the χ^2 is artificially decreased by a factor of almost 2.

9. Page 6, Equation (5) makes no sense in a Least Square retrieval approach, which seeks for a global minimum. Why one should look for a partial minimization, while using a simultaneous approach?

10. Page 7, lines 2 to 6. A χ^2 variable with $n = dim(Y)$ degrees of freedom has mean n and variance $2n$. Because for n large, the χ^2 distribution is approximately Gaussian, to compute a χ^2_{th} −tolerance limit, say within 3 standard deviations (or 3σ), we just need to calculate $\chi^2_{th} = n + 3\sqrt{2n}$. As an example, for $n = 103$, the number of channels the authors use in their B2 band, we have χ^2_{th}≈145, or $\frac{\chi^2_{th}}{n} \approx 1.42$. Conversely, the authors use $\frac{\chi^2_{th}}{n} = 4$, which in view of the factor 2 above (see point 8) increase to 8, which corresponds to a tolerance interval of 50 (fifty) standard deviation (sic!). With this convergence criterion, almost all retrievals are not converged!

11. Page 7, Equation (6). I do not like the use of this empirical *Contamination Factor.* Why the final solution should be contaminated? They use a simultaneous retrieval. Averaging Kernels are good to assess vertical resolution. To check the interdependency of the retrieved state vector the authors have the a-posteriori covariance matrix. Please, use this matrix and compute the correlation matrix. In case the N_2O profile has not been independently resolved, the authors will see a relatively large correlation with other parameters, e.g. H_2O. If so, they have only one way to go, change or improve the retrieval algorithm, e.g., by using more IASI channels, which are sensitive to H_2O but not to N_2O. You have a lot in IASI.

12. Page 7 to 10, from Section Validation. With broad AK and a peak of 0.2 at most, the information retrieval comes from the background. There is no point in assessing this layer-by-layer accuracy. All AK broadly peaks around 400 mbar, which means that the N_2O at this layer also receives contribution from the rest of profile. The authors need to check for correlation. In case, as I suspect, the N_2O layer-retrieval at about 309 is strongly correlated with the rest of the profile, the accuracy alone (root mean square error of the a-posteriori covariance matrix) is not a good quality index. I suggest that the author should also compute the N_2O total column. Because of integration along the vertical, this parameter will be more depending on the true state than the background.

13. Page 11, Section Troposphere variations….Since the authors failed to show that the N_2O layer-retrieval at 309 hPa is independently resolved, the results in this section could be seriously flawed. Once again, by looking at AK in Fig. 6 and 7 the 309 hPa layer retrieval of N_2O gets contribution from any other layer along the profile. It is important here that the authors show maps of the correlation matrix to get insight into a better understanding of the retrieval quality and accuracy at 309 hPa.

Minor Comments

14. Page, line 11. The authors say*, … Over the mid-latitude regions, both variations of N2O_B1 and N2O_B2 at 309 hPa are influenced by the stratospheric N2O-depleted air* ***because*** *of the relative coarse shape of the averaging kernel…*I have found clumsy sentences like this throughout the paper. It seems that the *cause* of a physical phenomenon is the mathematical structure of IASI AK. I think that authors here want to say that …*Because of the relative coarse shape of the averaging kernel, IASI is sensitive to variations of N2O_B1 and N2O_B2 at 309 hPa influenced by the stratospheric N2O-depleted air.* Here the authors seem to suggest that the best retrieval function capable of assessing this phenomenon could be a proper average over the broader part of AK, but then they go completely another way and focus on a single layer.

15. Page 3, section 2 IASI. Please put the reference to IASI at the beginning of the section….*IASI (Hilton et al 2011)*…Furthermore, on line 11 remove the reference to Clerbaux et al 2009, it is not appropriate once you have cited Hilton et al at very beginning. Furthermore, on line 13 the reference again to Hilton et al is redundant. Please, remove it.

16. Page 3, line 21, Change *RRTOV is a fast…* in *RTTOV is a polychromatic fast…*

17. Page 4, Beginning of section. That the IASI spectral coverage includes the N_2O ν_1, ν_3 fundamental absorption band is a well assessed result from molecular spectroscopy, please rephrase the sentence on line 2, the reference to Clearbaux et al 2009 is not appropriate and unnecessary.

18. Table 1 is never called in the text. Please make proper reference to this table in the text and show also the number of IASI channels used for retrievals.