

## Anonymous Referee #2

Received and published: 15 May 2018

This paper presents results from N<sub>2</sub>O IASI retrievals based on the RTTOV radiative transfer model. N<sub>2</sub>O satellite observations are important to understand its global distribution and maybe help characterizing its emissions. As mentioned below, IASI has already been used to retrieve N<sub>2</sub>O profiles and EUMETSAT retrieval algorithm is providing such data for the whole IASI period. Therefore, the present study is not providing completely new data. It is nevertheless interesting to have more than one dataset from the same instrument in as much as the quality of the datasets are proven. The objective of the paper is two-folded. The presentation of the retrieval methodology and validation and a case study. As detailed below the methodology and validation part should largely be strengthened and the Hysplit transport study which is weak could be removed.

Overall the quality of this study is not good enough to be published in AMT. I have major concerns about the originality, the methodology and the results that are presented in the paper.

### I-Originality of the work

There are other studies on IASI N<sub>2</sub>O retrievals which are not sufficiently acknowledged and discussed. One of the first publication about N<sub>2</sub>O IASI retrievals is Garcia et al. (AnnGeo, 2013). Based on one year of data they show that the N<sub>2</sub>O EUMETSAT v5 product (August et al., JQSRT, 2012) provides a good agreement with FTIR data at Izaña for the 10-14 km vmr. Garcia et al. (AMT, 2016) make a comparison between the EUMETSAT v5 product and the Izaña FTIR data for 4 years. These comparisons show a very good agreement (R=0.87) for the total columns annual cycle. In their latest paper Garcia et al. (AMTD, 2017) show a good agreement between IASI N<sub>2</sub>O and HIPPO data. The authors should use these previous studies in details rather than just citing them. In particular they should discuss and compare their retrieval methodology, characterization and results with those described in these papers throughout the manuscript.

→ Interannual trends and seasonality of N<sub>2</sub>O have been widely addressed by the retrieved data presented in the previous studies. This work aims to present a new retrieved N<sub>2</sub>O data which are of a sufficient quality to be used to study spatial and temporal variation of N<sub>2</sub>O on a daily basis. As it was demonstrated in Kangah et al., (2017), these variations could be footprints of high N<sub>2</sub>O emission hotspots especially over Asia. Thus, to enhance the originality of this work in comparisons to the other studies, we added some sentences in the introduction part of the paper as follows:

These operational N<sub>2</sub>O total column products also show seasonal cycles and annual trends consistent with the retrieved N<sub>2</sub>O from the ground-based Fourier Transform Spectrometer (FTS) observations at the Izaña Atmospheric Observatory (IZO, Spain) (García et al., 2014, 2016). First results of N<sub>2</sub>O total columns retrievals using a partially

scanned IASI interferogram with an accuracy of  $\pm 13$  ppbv ( $\sim 4\%$ ) are described in Grieco et al. (2013). Retrievals of N<sub>2</sub>O tropospheric profiles have been performed using the Atmospheric Infrared Sounder (AIRS) and the results showed global interannual trends consistent with surface measurements (Xiong et al., 2014). N<sub>2</sub>O profiles retrieved from the Greenhouse Gas Observing Satellite (GOSAT) measurements have been used to study the transport of Asian summertime high N<sub>2</sub>O emissions to the Mediterranean upper troposphere (Kangah et al., 2017). Using monthly averaged GOSAT N<sub>2</sub>O retrievals at 314 hPa together with outputs from the chemistry transport model LMDz-Or-INCA, this study evidenced the transport of high surface N<sub>2</sub>O emissions from Asia to the upper tropospheric Mediterranean during the summer monsoon period. This was the first study to link upper tropospheric N<sub>2</sub>O spatial and temporal variations to regional emissions hotspots seasonality using retrievals from satellite measurements. In this paper, we describe the IASI instrument and the Radiative Transfer for Tiros Operational Vertical sounder (RTTOV) used as forward model in our retrieval system in sections 2 and 3, respectively. We present the retrieval strategy and the validation of the results using HIPPO airborne in situ measurements in sections 5 and 6, respectively. In section 7, we analyse the scientific consistency of the retrievals focusing on the long-range transport of N<sub>2</sub>O during the Asian summer Monsoon. In this part, we show that the N<sub>2</sub>O transport processes between the Asian surface and the eastern Mediterranean demonstrated in kangah et al., (2017) can be followed using our retrievals at a finer timescale, namely on a daily basis.

Moreover, the consistency between our retrieval vertical sensitivity and the previous studies is detailed in the manuscript as follows:

In addition, the averaging kernels corresponding to this level peak at 309 hPa. Therefore, retrieved vmrs at this level are the most reliable for both N<sub>2</sub>O\_B1 and N<sub>2</sub>O\_B2. For N<sub>2</sub>O\_B2, all the averaging kernels peak at the levels 309 hPa. This means that the retrieved N<sub>2</sub>O vmr profiles are mainly sensitive to the real

N<sub>2</sub>O vmr at this level. This result is consistent with previous studies from Kangah et al. (2017), Xiong et al. (2014), Grieco et al. (2013) and Garcia et al. (AMT, 2018).

The following reference have been inserted in the revised version:

García, O. E., Schneider, M., Ertl, B., Sepúlveda, E., Borger, C., Diekmann, C., Wiegele, A., Hase, F., Barthlott, S., Blumenstock, T., Raffalski, U., Gómez-Peláez, A., Steinbacher, M., Ries, L., and de Frutos, A. M.: The MUSICA IASI CH<sub>4</sub> and N<sub>2</sub>O products and their comparison to HIPPO, GAW and NDACC FTIR references, *Atmos. Meas. Tech.*, 11, 4171–4215, <https://doi.org/10.5194/amt-11-4171-2018>, 2018.

Concerning the retrieval accuracy, we inserted the following paragraph in the revised version:

... MUSICA/IASI retrieved N<sub>2</sub>O (Garcia et al., 2018) presented a R<sup>2</sup> (0.22) nearly similar to that of N<sub>2</sub>O\_B1 (0.18) but with a greater std error (~2.5 %). Comparing with HIPPO, GOSAT N<sub>2</sub>O retrievals (Kangah et al., 2017) have a std error of about 0.6 %, a R<sup>2</sup> of about 0.19 and a slope of about 0.22. In addition, MUSICA/IASI retrieved N<sub>2</sub>O have a degree of freedom of about 1.39 which is nearly the same than the one of N<sub>2</sub>O\_B1 (1.38).

These results must be compared very carefully with those of the present paper since there are significant differences in the error analysis strategy and reference datasets between the different studies. Thus, in the one hand, MUSICA/IASI retrieved N<sub>2</sub>O are compared with HIPPO datasets using a smaller number of collocated pixels (N=23) than that we used (98 and 102 collocated data for N<sub>2</sub>O\_B1 and N<sub>2</sub>O\_B2, respectively) and in the other hand, GOSAT N<sub>2</sub>O retrievals have been validated only over maritime pixels. Since the linear regression is very sensitive to this kind of differences, we can only assess a very qualitative and approximative comparison between these retrievals. Therefore, we can consider, at first glance, that MUSICA/IASI retrieved N<sub>2</sub>O seems qualitatively consistent with N<sub>2</sub>O\_B1 and GOSAT N<sub>2</sub>O in terms of accuracy and vertical sensitivity.

To assess an exact and detailed comparison between the different kind of retrieved N<sub>2</sub>O, an inter-comparison study must be performed. This is out of scope of this paper which aims to present a new retrieved N<sub>2</sub>O results with their strength and weaknesses and the kind of scientific study these new N<sub>2</sub>O products can be used for.

IASI-A is flying since 2006 and the present paper presents retrievals for validation with HIPPO data and a series of situations over a limited region for a very limited time period. It is possible to accept such a limited study for a very recent mission but difficult for a ten years mission with previous studies much more extended already published. Indeed, as mentioned above, in their latest studies Garcia et al. have taken advantage of the long time series to make robust statistics and they have used different available validation datasets such as long term FTIR profiles and columns at Izana and GAW in-situ data (see reply to reviewers in Garcia et al.,

AMTD 2017) and HIPPO campaigns. The present paper would be more convincing if it could prove that the new IASI N<sub>2</sub>O retrievals provide robust information about the N<sub>2</sub>O variability taking advantage of the large IASI period which is not yet the case.

→ The HIPPO campaigns cover the 4 seasons and almost all the latitudinal bands (cf. figure 4 of the manuscript). There are therefore, at this time the best database to validate N<sub>2</sub>O retrievals. In addition, our aim (cf. response #1-) is to demonstrate that our data are useful to study spatial and temporal variations of N<sub>2</sub>O on a daily basis. Our strategy here is to take advantage of the knowledge of the summertime N<sub>2</sub>O transport processes between the Asian surface and the Mediterranean which have been assessed on a monthly basis in the previous study of Kangah et al., (2017) and show that our retrievals can capture this transport with finer timescales. For this purpose, we don't need to use all IASI data since 2006.

## II-Methodology:

The retrieval methodology is not fully presented and justified. In page 5 the basic equation of the OEM are rewritten which is unnecessary. They are described and explained in Rodgers (2000) and many other publications and can therefore be removed and replaced by more interesting information. Indeed, the retrieval strategy itself is hardly described and justified. Many auxiliary parameters are retrieved together with the absorbing gases profiles but no justifications and no discussion about these retrieved parameters are given.

→ We clarified the retrieval methodology in the revised version of the manuscript (cf. responses #2, #3, #4, #8 to the referee #1). Concerning, the auxiliary parameters, the figure 2 of the manuscript highlights the key parameters in each band which should be accurately known to perform good N<sub>2</sub>O retrievals. In addition, error covariance matrix used for these by-products are also described and justified. Furthermore, the last important thing about these parameters is to estimate and as far as possible to remove their impact on the N<sub>2</sub>O retrievals. This is what we did by using the Contamination Factor (see #II-i).

i-Contamination Factor: This part is interesting because it allows to document how uncertainty on an auxiliary parameter will impact the retrieved target state vector. Nevertheless it is only valid for auxiliary parameters that are kept constant and are not retrieved together with the target parameters. In case of retrieved parameters, it only gives an idea of the parameters which retrieval will mostly interfere with the target parameters but does not allow us to know the quantitative impact on the target parameters. The authors should explain that this methodology is not quantitative for retrieved parameters.

→ We do not agree with the referee concerning the Contamination. In a simultaneous retrieval strategy, we can also assess how uncertainty on a co-retrieved parameter will impact the target species (see response #11 to the referee #1).

ii-Atmospheric temperature retrieval: Why is the atmospheric temperature retrieved together with N<sub>2</sub>O and the other interfering species? Where is the information about the atmospheric temperature profile coming from? Atmospheric temperature is normally retrieved from CO<sub>2</sub> lines assuming constant CO<sub>2</sub> vmr's. CO<sub>2</sub> or other gases vmr's are retrieved assuming constant atmospheric temperatures. These procedures avoid mixing between T and gases retrievals.

Here there are some CO<sub>2</sub> lines in the B2 band but the most likely is that the temperature is retrieved from other absorption lines such as N<sub>2</sub>O. The risk of contamination and interference is therefore major. This is actually shown by figure 5 and 6 where the CF are drawn. Atmospheric temperature uncertainties have the largest impact on N<sub>2</sub>O retrievals in both B1 and B2 with CF a factor of 4 or more larger than for the other parameters in the mid-upper troposphere. As stated above, this means that the T and the N<sub>2</sub>O retrievals are not independent. Therefore high (low) N<sub>2</sub>O could be caused by high (low) T or the other way around but the impact cannot be determined because of the joint retrieval.

→ Both B1 and B2 are sensitive to the temperature which the prior knowledge is from the operational level 2 products. Thus, we can either fix the temperature profile to these a priori profiles or co-retrieve the temperature profiles simultaneously with N<sub>2</sub>O. This latter strategy that we used in our study allows a global adjustment of the calculated radiances together with all the parameters that the forward model is sensitive to. However, the two retrieval strategies allow to assess the contamination on the target species. When the interfering parameter is fixed, the contamination can be quantified via the model parameter error (cf. Rodgers, 2000 page 48) and when it is part of the state vector it can be quantified via the extra-diagonal elements of the global averaging kernel matrix (cf. Rodgers and Connor 2003; Rodgers, 2000 page 70) as it is explained in the response #11 to the referee #1. The CF of T is larger than the CF of H<sub>2</sub>O. However, since the variabilities of water vapor and therefore of its CF are larger than for the other parameters, the CF of H<sub>2</sub>O results in a higher impact on the N<sub>2</sub>O spatial and temporal variability especially over tropics. The figures 11, 12 and 14 clearly show this impact over tropics. Thus, this difference of behaviour between N<sub>2</sub>O\_B1 and N<sub>2</sub>O\_B2 especially over tropics shows that the parameter which impact the N<sub>2</sub>O variability is not T which CF is similar in B1 and B2.

iii-Emissivity retrieval: The authors state that in RTTOV the ocean emissivity is parameterized and that land emissivity is prescribed by an atlas. They call these emissivities a priori emissivities and retrieve surface emissivity in their procedure. Are the emissivities spectrally varying in RTTOV? How are the emissivity jacobians computed in RTTOV? Are they the same over sea and over land? It would be interesting to see results from emissivity retrievals and the differences over sea and land and over different types of land. Surface temperature and surface emissivity are parameters with signatures hard to discriminate in a small spectral window such as B1 or B2 as they basically give the background slope. The retrieval of both parameters is probably redundant. The authors should give information about how much the spectral chi-square has been improved when surface emissivity is retrieved and about the improvement it provides on the validation dataset. In case of no or too small improvements, the retrieval procedure has to be reconsidered without emissivity retrieval.

→ In RTTOV, surface emissivity over land and ocean are spectrally varying. The computation of the emissivity Jacobian is similar as for the other parameters. It is computed by analysing the perturbation of the forward model for a given perturbation of the parameter assuming a linear relationship between these two perturbations ( $\delta\mathbf{y} = \mathbf{H}(\mathbf{x}_0)\delta\mathbf{x}$ ). It is true that the spectral information of the surface temperature and the emissivity are difficult to discriminate. In our retrieval, the surface temperature gives the most reliable physical information about the surface since the a priori profile and errors are taken from the IASI level 2. Thus, in our retrieval strategy, the emissivity behaves as a mathematical adjustment parameter since we took a

relatively high (~10%) a priori std errors. In addition, according to the figure 2, the spectral impact of the surface parameters (represented by surface temperature) is limited (but not negligible). The emissivity is therefore simply used as a by-product in our retrieval strategy and is not dedicated to being scientifically analysed as a retrieval product.

In addition, the value of the cost function  $\chi^2$  (which is neither a necessary and nor a sufficient condition for the quality of a retrieval) does not vary significantly with and without the retrieval of the emissivity. The spectral variation of the surface emissivity here contributes to stabilise the inversion scheme by mathematically “absorbing” some spectroscopic transition which are not accurately taken into account in our fast-radiative transfer model. This induced more realistic and stable vertical profiles. But this kind of methodology is very empirical and is hard to proved analytically.

iv-Validation: Equation 10 is applied to the HIPPO profiles to take the IASI vertical resolution and the impact of the a priori profile into account. Nevertheless, in order to apply this equation, the validation profiles have to cover the whole atmosphere. How and with what data are the tropospheric HIPPO profiles completed above the aircraft profiles top? How is the tropopause altitude taken into account? Concerning the comparison between the empirical and the theoretical errors there is a conceptual error. The authors compare the standard deviations of the differences between smoothed validation profiles and retrieved profiles (Emp) to the theoretical error (sum of smoothing and measurement errors Theoret) (Fig. 4). But as the validation profiles are smoothed by equation 10, the smoothing error is already taken into account and Emp has to be compared to RetNoise. As RetNoise is larger than Smooth this would not make a big difference. The other way is to compute the differences between the retrieved profiles and the raw validation profiles and to compare Emp with Theoret. Furthermore, the authors have shown that T uncertainty is largely impacting N<sub>2</sub>O retrievals (see CF) but as they retrieve jointly both parameters they cannot compute the resulting error. If the T profile was kept constant as suggested above, the errors caused T uncertainty could be evaluated (see Rodgers 2000). The errors caused by the other parameters should also be taken into account to compute the Theroret error but the same problem arises. The authors compute the  $S_e$  matrix to provide the smoothing error instead of using  $S_a$ . Nevertheless  $S_a$  should represent the actual N<sub>2</sub>O global variability as accurately as possible and is the matrix that should be used to compute the smoothing error in equation 7 (Rodgers 2000).  $S_e$  computed from the HIPPO data is representative of oceanic N<sub>2</sub>O profiles for given periods and may underestimate the variability. If the authors think it is a better representation of N<sub>2</sub>O global variability they have to justify this choice and may use it also for the retrievals. Furthermore a graphic representation of  $S_e$  (diagonal values and covariance/correlation) should be given and compared to  $S_a$ .

→ The HIPPO profile has been extended using the chemistry transport model LMDz-Or-INCA. This has been clarified in the revised manuscript:

Using a similar method as explained in Kangah et al. (2017), we used for these comparisons the measurements from the Harvard/Aerodyne Quantum Cascade Laser Spectrometer (QCLS), one of the airborne instruments of HIPPO, and the retrieved profiles selected within a collocation temporal and spatial window of ±200 km and ±12h,

respectively. We extended the HIPPO profiles using monthly averaged profiles from the chemistry transport model LMDz-Or-INCA. To minimize the impact of this extension and since we are interested in the upper tropospheric N<sub>2</sub>O (cf. paragraph 6.2), we only took HIPPO profiles with a ceiling pressure less than 250 hPa and a bottom pressure greater than 400 hPa.

The tropopause altitude is then considered via the model.

There is no conceptual error concerning the comparison between the empirical and the theoretical errors. The retrievals are compared with the raw HIPPO profiles to estimate the empirical errors. So, the empirical error can be compared with the sum of the smoothing error and the retrieval noise. This has been clarified in the revised manuscript:

The theoretical covariance matrix of the total errors is then compared with an empirical total errors covariance matrix calculated using the raw (without applying the retrievals averaging kernels) HIPPO measurements and the retrievals along the HIPPO campaigns flight paths (namely the covariance matrix of the difference between HIPPO profiles and IASI retrieved profiles).

The theoretical basis of the CF has been explained in II-i and II-ii.

Concerning the use of  $S_e$  instead of  $S_a$  for the smoothing errors, Rodgers (2000, page 49) explained the necessity of using a matrix which is more accurate than  $S_a$  to compute the smoothing errors. Since, the smoothing errors should also represent the loss of fine structures, a statistic of these fine structures must be used. Thus,  $S_a$  which is a reasonable constraint for the retrieval can be not enough accurately build to describe the statistics of these fine structures. That why we used the HIPPO in-situ profiles to build  $S_e$  and estimate accurately the smoothing errors of the retrievals. HIPPO is mostly over the ocean but have a significant number of profiles over land. In addition, HIPPO N<sub>2</sub>O database gives the whole N<sub>2</sub>O tropospheric profiles both latitudinal variations (one of the dominant variation mode) and seasonal variations through the 5 HIPPO campaigns. Of course,  $S_e$  is not perfect and maybe slightly underestimates the variations of N<sub>2</sub>O over land surface.

$S_e$  and  $S_a$  are therefore different matrices playing different roles in the retrieval process and characterisation. We can't use  $S_e$  as apriori error covariance matrix because that will induce a dependency between the retrieval results and the validation data.

Instead of  $R$  we should have  $r^2$  which shows the percentage variation in the retrieved profile that is explained by the variations of the validation profile. Therefore  $R > 0.707$  is needed to have more than 50% of the retrieved variability coming from the real variability. It is also important to have a comparison of the variability of the validation data and of the retrieved data. All this information (standard deviation of the differences,  $r^2$ , variability) should be given synthetically with a Taylor diagram.

→ We agree with the reviewers concerning the fact that  $R^2$  should also be given, since it shows how much the linear regression with the reference dataset explains the distribution of our retrieval. So, we also mentioned this parameter in the revised version:

... N<sub>2</sub>O\_B1 and HIPPO measurements are moderately correlated (the Pearson linear correlation coefficient  $R=0.42$ ) with a low bias and standard

deviation (called hereafter std) error of -1.6 ppbv (~0.5%) and 3.5 ppbv (~1.0%), respectively. Thus, the linear regression using HIPPO measurements explains 18% ( $R^2=0.18$ ) of the variations of  $N_2O\_B1$ .

...

The consistency between  $N_2O\_B1$  and HIPPO increases at mid-latitudes (e.g.  $R^2=0.4$  for northern hemisphere mid-latitudes). We can also notice that there is a very low mean bias (-0.1 ppbv) in the northern hemisphere high-latitude regions.

...

$N_2O\_B2$  is moderately correlated with HIPPO measurements ( $R=0.6$ ) with a std error of 3.2 ppbv and a very low mean bias of 0.3 ppbv. Thus, 36% of  $N_2O\_B2$  variations are explained by the linear regression with HIPPO measurements.

...

In tropical regions, the correlation coefficient between  $N_2O\_B2$  and HIPPO measurements becomes very high (0.71 and 0.92 in the northern and southern hemispheres, respectively) compared to the other regions. Therefore, in tropical regions more than 50% of  $N_2O\_B2$  variations are explained by the linear regression with HIPPO measurements.

The figures 9 and 10 show scatter plots of our retrievals and HIPPO in-situ  $N_2O$  measurements and synthetically give informations about the systematics errors (bias), the random errors (std error) and the accuracy of the variations ( $R$  and regression slope). We also give the variation explained by the linear regression with the reference datasets ( $R^2$ ). This kind of plots are largely used in retrievals validation studies and therefore allow a quick comparison between different retrieval datasets. We think that there is no need to use a Taylor diagram here.

### III-Results:

i-Validation: The retrieval results are not fully convincing. When the whole HIPPO dataset is used, meaning the strongest statistics ( $N$  about 100),  $r^2=0.18$  for  $B1$  and 0.36 for  $B2$  implying only 18 and 36% of the retrieved variability explained by the actual variability. Even if based on a limited HIPPO dataset, Garcia et al. (2017) achieve a better correlation ( $r^2 = 0.58$ ) with a similar type of comparison as presented here. As they deal with a very close type of comparison, the results of Garcia et al. (2017) even in a paper under review should be discussed here. In most latitudinal bands (weaker statistics with  $N < 30$ )  $r^2$  is lower than 0.5 especially in the  $B1$  case with a maximum of 0.4 in the northern mid-latitudes. In the  $B2$  case  $r^2$  is the highest (0.85) for the tropical southern latitudes. But in that case it is based on 12 points only which makes the statistics really poor and the high  $R$  is due to the fact that the points are separated in to clusters. Furthermore, in the best  $r^2$  cases (tropical southern and northern latitudes for  $B2$ ) the slopes of the linear interpolation are much larger than unity (2.5 and 3.3) indicating a largely too strong variability of the retrieved vmr's compared to the validation vmr's. For northern mid-latitudes  $r^2 = 0.4$  for  $B1$  and 0.29 for  $B2$  which are rather low values. Finally, the authors state that in summary  $N_2O\_B1$  and  $B2$  are of sufficient quality to analyse  $N_2O$  variations in the mid and high latitude regions. This conclusion is not really supported by the validation results as discussed above. Especially for high northern latitudes with  $r^2= 0.1$  for both  $B1$  and  $B2$ , only 10% of the variability comes from the actual  $N_2O$  variability. We would rather say that these data should not be used.

→ We do not agree with the referee concerning the quality of the retrieval. For N<sub>2</sub>O\_B1, at global scale R<sub>2</sub> is about 0.18 (merging all latitudinal bands) and about 0.4 over mid-latitude regions. These results are consistent with those presented in Kangah et al., 2017 for GOSAT N<sub>2</sub>O retrievals maritime pixels and in Garcia et al. (2018). In addition, from the comparison with HIPPO, N<sub>2</sub>O\_B1 present better accuracy (bias and  $\sigma$ ) than the previous studies (Xiong et al., 2014; Garcia et al., 2018; Kangah et al., 2017). Results from Xiong et al., has been successfully used to derived global trends of N<sub>2</sub>O and GOSAT Mediterranean N<sub>2</sub>O have been linked to high N<sub>2</sub>O emissions hotspots over Asia. This means that having a R<sub>2</sub> < 0.5 does not mean that the data are not good enough to be used for scientific purpose. Therefore, our current N<sub>2</sub>O\_B1 product can at least be used for this kind of studies.

In addition, for N<sub>2</sub>O\_B2 the results of the comparison with HIPPO are better than all current validated N<sub>2</sub>O profiles products in terms of R, R<sub>2</sub>, std errors and bias. Thus, N<sub>2</sub>O\_B2 can be used at finer time scale especially in tropical regions where R<sub>2</sub> is better than 0.5 (R<sub>2</sub>=0.5 for northern hemisphere tropical regions over N=32 collocated pixels).

ii-Transport study: The variability of IASI N<sub>2</sub>O at 309 hPa shown on Fig. 13 is probably coming from a tropopause height difference. As shown by the AvK's, IASI vmr at 309 hPa is sensitive to a very large altitude range (600-120 hPa). Therefore it is equivalent to a N<sub>2</sub>O column or mean vmr over this range. When the tropopause changes from ~100 hPa in the tropics to ~250 hPa in the extratropics, the corresponding N<sub>2</sub>O columns mechanically change because the N<sub>2</sub>O vmr is lower in the stratosphere than in the troposphere. The authors attribute the N<sub>2</sub>O enhancement to upward transport from the Asian BL and horizontal transport within the anticyclone. This is also probably the case as shown by an extended literature based on satellite CO observations (Park et al., JGR, 2007...). Nevertheless, N<sub>2</sub>O is a well mixed gas and the quantification of such an effect is rather complicated. Surface in-situ data generally show a very limited seasonal variability of the N<sub>2</sub>O mixing ratio even in emission regions. Therefore the Asian BL is probably not N<sub>2</sub>O enriched as it is CO enriched. If the authors have evidence and data to document an important N<sub>2</sub>O enrichment during the monsoon in south Asia they should provide and discuss it. Another element that tends to strengthen the tropopause effect is that the IASI N<sub>2</sub>O high values are not limited to the anticyclone boundaries but to the whole tropical region. See in particular the high N<sub>2</sub>O band between 15 and 5°N which is outside of the anticyclone (the southern boundary of the anticyclone is at about 15°N). In order to have a better idea of the tropopause versus BL transport effects (i) the region of Fig. 13 should be extended both in latitude and longitude (ii) the boundaries of the anticyclone should be provided on Fig. 13 based for instance on PV values (see Ploeger et al., ACP, 2017) or on geopotential height values (e.g. Randel and Park, JGR, 2006). The Hysplit study is based on online simulations and simply shows that on the southern edge of the anticyclone, transport is westward which is expected. It does not prove that the air parcels are coming recently from the south Asian BL (the backtrajectories end up between 700 and 300 hPa and with a tenths of trajectories the statistics are very poor when Lagrangian studies are performed with millions of air parcels) nor that N<sub>2</sub>O enhancements over the whole tropical band could be due to such a transport process. The Hysplit part is therefore largely insufficient to draw conclusions and could be removed. The literature is rich enough about the subject of upward transport of BL air masses to the UTLS and trapping of pollution into the anticyclone. See for instance the Lagrangian modeling study of Bergman et al. (2013). References to this extended literature are enough.

→ The emission and the transport of N<sub>2</sub>O from Asian BL to the Mediterranean Basin during the summer monsoon period has been largely addressed in Kangah et al., 2017. These high emissions during summer due, among others, to the high soil water content can be observed in current N<sub>2</sub>O emissions cadastre (e.g. cf. Kangah al., 2017, fig 9). The part 7 of the manuscript aimed to show that the spatial and temporal variation of N<sub>2</sub>O\_B2 is consistent with this proved long-ranged transport structure despite the smoothing effect due to the shape of the averaging kernels (smoothing errors) and the retrieval noise. Thus, over the period 21-23 July for instance, the figure 15 show a relatively homogenic tropopause level where the figure 14 show spatial variations of N<sub>2</sub>O over Asia with hotspots over eastern China and the Indian-Tibetan Plateau regions which were expected from the literature. This cannot be explained simply by the tropopause effects. The high N<sub>2</sub>O emissions are observed in most south Asian regions with hotspots over India and the north-eastern China. Thus, high N<sub>2</sub>O vmr are also expected in upper troposphere in all these regions in addition to the accumulation effect due to the monsoon anticyclone.

We agree with the reviewer with the fact that the hysplit part is not enough to draw conclusions. We used this part as an additional building block in our demonstration not as a self-sufficient assessment. This part is interesting, as it shows that the westward transport from Asia to the Mediterranean is consistent with daily N<sub>2</sub>O transport fluxes as represented by N<sub>2</sub>O\_B2.

#### IV-Minor comments:

p2 I20-29: To my knowledge, the first paper to deal with tropospheric N<sub>2</sub>O retrievals from a satellite instrument is Chedin et al. (GRL, 2002). It shows very interesting results concerning the N<sub>2</sub>O evolution based on the TOVS instrument. This ref should be cited in the paper.

→ We have inserted a sentence relative to these observations.

Chédin et al. (2002) show the annual and seasonal variations of N<sub>2</sub>O concentrations retrieved from the Television and InfraRed Operational Satellite-Next generation (TIROS-N) Operational Vertical Sounder (TOVS) instrument.

Chédin, A., Hollingsworth, A., Scott, N. A., Serrar, S., Crevoisier, C., and Armante, R., Annual and seasonal variations of atmospheric CO<sub>2</sub>, N<sub>2</sub>O and CO concentrations retrieved from NOAA/TOVS satellite observations, *Geophysical Research Letters*, 29(8), 2002.

p3 I16: Turquety et al. (2004) does not concern IASI O<sub>3</sub> retrievals. There are a number of recent refs concerning IASI O<sub>3</sub> retrievals.

→ Done. We added the following reference in the revised manuscript:

Dufour, G., Eremenko, M., Griesfeller, A., Barret, B., LeFlochmoën, E., Clerbaux, C., Hadji-Lazaro, J., Coheur, P.-F., and Hurtmans, D.: Validation of three different scientific ozone products retrieved from IASI spectra using ozonesondes, *Atmos. Meas. Tech.*, 5, 611-630, <https://doi.org/10.5194/amt-5-611-2012>, 2012

P4 I17-18: the authors should give a recent reference to justify their choice of NEDT.

→ Done.

We used for our retrievals NEDT from Clerbaux et al., 2009.

P5 I16: the authors should give a ref or a detailed explanation that justify the shape of their a priori covariance matrix. We also need information about the shape of the a priori matrices for the other retrieved profiles (are they diagonal?).

→ The a priori covariance matrix is derived and adapted from Rodgers, (2000, eq. 2.83). Concerning the shape of the whole matrix the manuscript has been modified as follows:

The a priori error covariance matrix  $S_a$  is built for all chemical species and by considering parameters independent to each other as follows (cf. Rodgers, 2000):

$$S_{aij} = \sigma_a^2 \times \exp(-|\ln(P_i) - \ln(P_j)|) \quad (3)$$

where  $\sigma_a^2$  is an a priori variance error fixed for each parameter of the state vector and  $P_i$  the pressure level at the level  $i$ .

Diagonal matrices are used for temperature profile and surface emissivity.

P6 I2: the choice of 30% for the a priori variability for H2O because of HDO is rather empirical and poorly justified. What does sink parameter mean?

→ cf responses #7 and #11 to the referee #1.

P6 I4 and I6: sensitivity studies are mentioned but the reader knows nothing about what they are made of. Details about the methodology used and about the results of these sensitivity studies are needed.

→ We removed this unclear expression since we fixed the a priori std errors using the estimated current knowledge about the variations of each parameter. The sensitivity studies are done via the figure 2 to decide which parameter should be retrieved or not.

P6 I14: ref for the radiometric noise (see above).

→ Done

Figures: Fig 14: this figure is of poor quality and should be improved. The winds should be superimposed such as on Fig. 13 in order to make a more straightforward comparison.

→ Done