In this study, the authors investigate the effects of gravity wave breaking and the resulting turbulence on mixing around the tropopause. The study is based on a research flight over the Southern Alps during the DEEPWAVE measurement campaign. In-situ measurements of N2O and CO above and below the tropopause, upstream and downstream of the mountains, have been used to diagnose mixing, while gravity waves and turbulence were analysed in detail using temperature and wind measurement data. The authors report breaking gravity waves and air turbulence close to the tropopause, and a resulting alteration in tracer structure, which shows that significant mixing events have occurred in the affected atmospheric regions.

Overall, I believe the results presented in the manuscript are of a very high standard and clearly merit publication in ACP. The effects that gravity waves have on atmospheric composition and dynamics are still poorly understood, but of high relevance for quantifying tracer transport and large scale dynamics of the atmosphere. The main topic of the paper is therefore highly relevant and of considerable interest to the community. The methods and analysis in this work are generally solid and clearly presented, the analysis of cross-isentropic transport is especially detailed. The figures are well prepared. Presentation of results is also very clear in most parts, I would only suggest to make the mathematical notation more consistent in a few places and to clarify the identification of different flight segments discussed in the text (see minor/technical comments).

Authors response:

We thank the reviewer for the careful reading and suggestions which helped to improve the paper. We hope that we adequately addressed the key points. The reviewer comments are given in black, our comments are given in blue, text changes in the manuscript in red.

My two more substantial observations are given below, followed by a list of technical corrections and minor suggestions.

General points and related observations:

1) The UTLS region is known for sharp tracer gradients, both horizontal and vertical. The horizontal length scales of tracer filaments resulting from various stratosphere-troposphere exchange (STE) processes (like, for example, planetary wave breaking) can be much smaller than the dimensions of the flight pattern considered here. The structure of N_2O distribution in UTLS, as described in this paper, is shaped by STE and can be affected by various STE events. Since the speed of the aircraft is much larger than that of the wind, the air masses sampled downstream of the mountains are most likely not the same air masses as sampled upstream (or are they at least partially the same?), and thus it is possible that these air masses already had different N2O-θ profiles even before crossing the mountains. It would be good if authors could comment on such a possibility, or argue why it would not significantly alter the tracer gradient analysis results. It was briefly mentioned that aircraft was not flying through tropopause folds, but air with altered tracer structure could have been advected from elsewhere. I realise that this problem may indeed be very hard to address using only data from airborne in-situ measurements, but there are other arguments that could be made. For example, model data showing no complex structures in the typical stratospheric or tropospheric tracers upstream of the mountains before the flight could strengthen the argumentation that leads to the main conclusions of the paper. N2O data would, of course, be best, but ozone and water vapour, which should be available from ECMWF IFS, could already tell a great deal about possible influence of earlier STE events on the observed air masses. Alternatively, dynamical histories of the sampled air parcels and their surroundings could be investigated. There are also a few interesting details in the manuscript that might make this point more relevant:

Authors response:

We thank the reviewer for this point, which is central for our analysis. We checked ERA5 reanalysis data for ozone along the flight track in isentropic coordinates at 15:00 UTC (4 hours before our flight). As evident from the ozone cross section one can see, that a) the ozone distribution is almost isentropic, particularly at 330 K and that b) the vertical extent (in isentropic coordinates) of the ozone layers is rather constant upwind the islands. At 18:00 UTC at the time of our measurements ozone at 330 K is almost isentropically distributed. We therefore think, that advection of tracer gradients at 330 K are highly unlikely to have caused the different tracer gradients up- and downwind the mountains at 330 K.

Changes in manuscript:
The dynamical structure is mirrored by the ozone distribution from ERA5 data (not shown) along the flight track, which show a rather homogeneous distribution at Θ = 330 K (approximately flight altitude) notably upwind the region of our measurements.
a) The mechanism for modification of N2O-θ relationship by cross-isentropic transport and mixing, as described in Figure 8,

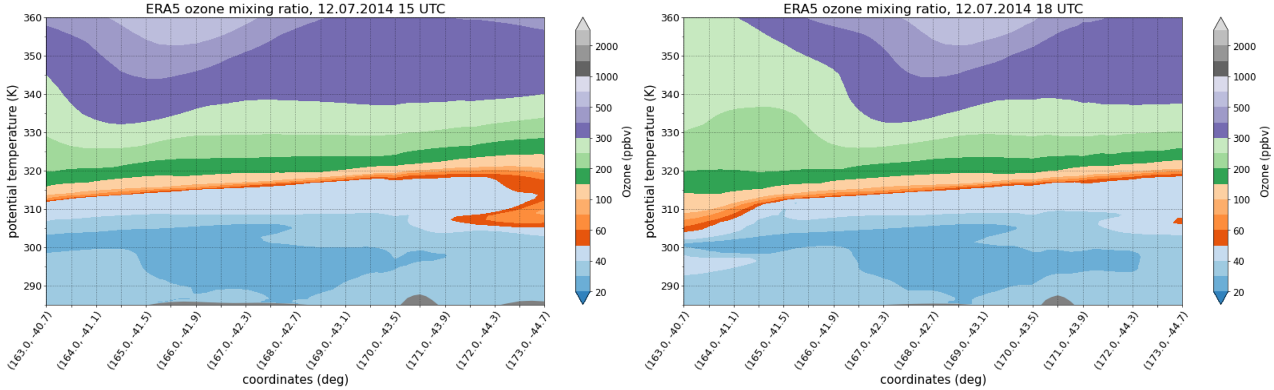

Figure 1. Vertical cross section of ozone from ERA5 along the axis of the flight track of interest for 15:00 UTC (left) and 18:00 UTC (right). Note the almost homogeneous ozone distribution at Θ = 330 K.

predicts that air above the mountains should include air masses that fall in between the compact upstream/downstream relationships (shaded region in Figure 8b). Therefore, in Figure 9, one would expect to see some black points (observations over the mountains) in between the compact relationships in blue and red (upstream and downstream data). However, the upstream data forms a compact relationship quite distinct from all the remaining higher-altitude flight leg data, especially in the 316-320 ppbv N2O range. Could this be a possible indication that some of the upstream air masses might have a different composition than over-the-mountain/downstream air masses had before being affected by GWs?

Authors response:
The reviewer is right with the observation, that at higher isentropes the N_2O-θ relationship differs. If we assume adiabatic flow in the upwind region, which is reasonable for stratospheric conditions within a short time period (hours) the air mass from higher isentropes would not affect the slope change at lower isentropes. Fig. 8 *(former Fig. 9)* shows that the slope between θ = 328 K to θ = 334 K changes from upwind to downwind in the region of orographic wave occurrence. The correlation of both air masses colored by the upwind, downwind above-mountain region consistent with the profile (Fig. 8 *(former Fig. 9)*, as suggested further below (see Fig. reply) highlights, that the composition between the tracers is almost the same upwind and downwind. This is, however not the case for the vertical profiles of CO and N_2O with respect to Theta (see CO profile). Turbulent mixing changes the tracer-Theta relation, with only weak effect (if at all) on the correlation. The fact, that there are no black points scattered between the upwind and downwind distribution might arise a) from the fact, that mixing may be incomplete not having homogenized the mixing ratios from the isentropes involved and b) the varying fractions of high-N_2O from lower isentropes and low-N_2O air from higher isentropes contributes and c) the flight track cannot cover all mixing states and regions with different mixing efficiency. The relative relation between the air masses indicates, that above the mountains the higher N_2O is more prominent at the flight levels compared to the downwind region.

b) Section 4.3 and the conclusions state that certain features of the results "can be seen as the result of the turbulence occurring potentially previously on this level". If the results suggest that the composition of (at least some of) the observed air masses was significantly affected by the previous turbulence/mixing processes, would it not be natural to ask if all the observed air masses were affected equally?

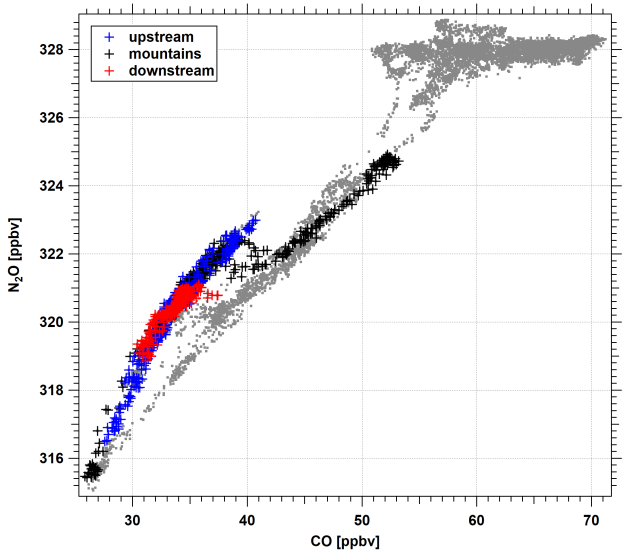

Figure 2. Scatter plot of N_2O vs CO for flight FF09 color coded according to the upwind, downwind and above-mountain regions as provided in the manuscript.

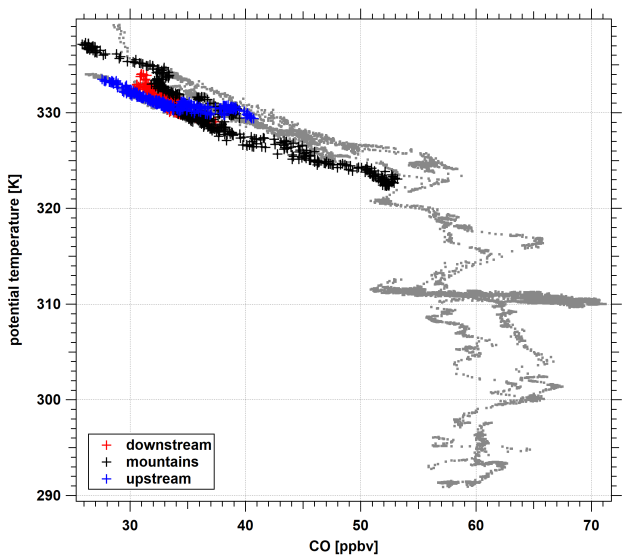

Figure 3. Profile of CO for flight FF09 (gray) as a function of Θ color coded according to the upwind, downwind and above-mountain regions as provided in the manuscript.

Authors response:

Similar to our comment above we have to accept, that we didn't sample the complete region of mixing. Note further, the transience of the processes: we might have missed the most active time of turbulence occurrence and mixing at the upper flight level. The fact that we observe only weak dynamical indication of turbulence above the mountains, but the slope change compared to the upwind relation is only explainable if we have a mixing processes between the upwind and downwind side. The further downwind, the stronger the effect of the mixing on the tracer distribution. This is illustrated at the lower flight level

(Fig. 3 *(former Fig. 4)*) which indicates turbulence occurrence at the flight track above and downwind the mountains, which leads to ongoing mixing also downwind the mountains.

2) After a dynamical process, such as wave breaking, causes cross-isentropic transport and scale breakdown in the tracer structure, tracers are further (mostly isentropically) mixed by molecular diffusion (e.g. Balluch and Haynes, 1997). The N2O-θ relationship is a great tool for characterising the cross-isentropic transport, but it would also be interesting to see to what extent the air masses that were transported across isentropes are already mixed into surrounding air. Maybe analysing the different air parcel groups from Figure 9 in N2O-CO space could shed some light on that? Or was N2O-CO analysis inconclusive for these air masses?

Authors response:
We provided the figure as suggested in the comment above. As can be seen, the distribution confirms the conclusion showing that the part of the flight track above the mountain has contributed to the mixing region (black dots scattered between the main correlation branches).
Two other findings are remarkable:
1) Also the downwind part shows signs of mixing (red data points between the main branches), which is inline with the behavior at the lower flight level shown in Fig. 3 *(former Fig. 4)*, illustrating high variability downwind the mountain.
2) The downwind correlation data appear as part of the upwind correlation data. At first this seems to be puzzling, but noting, that both $\mathrm{N_2O}$ and CO can be regarded as passive tracers, the relative relationship between both tracers should not be changed, when mixing occurs (turbulence acts in the same way to CO and $\mathrm{N_2O}$). The fact, that cross-isentropic mixing occurred (i.e. changing the tracer-Θ-relation) is untouched by this.

a) Another interesting feature of Figure 9 is that although the downstream air masses occupy roughly the same range of potential temperatures as the upstream ones, they have a much narrower range of N2O concentrations (close to the mean N2O value) with no outlying points in the rest of the upstream N2O range. Could this potentially suggest that the turbulence over the mountains, which the downstream air masses have experienced for just a few hours, has already mixed the affected air masses quite efficiently, and further isentropic mixing (which would normally be slower) is less relevant here? Again, maybe N2O-CO relationship could be used to confirm this?

Authors response:
Indeed we think, that the turbulent mixing is much more efficient and later (isentropic) mixing acts on longer scales. Inspecting the $\mathrm{N_2O}$-CO relation one could argue that turbulent mixing perturbs the canonical background correlations (as evident by the scatter data point between the main branches in Fig. 5 *(former Fig. 6)* (green) of the original manuscript). The isentropic mixing provides the background branches, which indeed are the result of mixing, but at different regions and on different timescales. However, this is a qualitative argument, but the fact, that those data points, which corresponds to the turbulence occurrence above the mountains and the different tracer-Θ relations upwind downwind clearly indicate an efficient diabatic (cross-isentropic) mixing process at the flight acting on the background distribution.

Minor and technical points:

Comment Page 1 Line 21: The phrase "conserves the effect" is confusing. It is not quite clear to me how an effect itself (as opposed to physical quantities or the results of the effect) can be conserved. This should perhaps be rephrased.

Reply to comment: We clarified the sentence. Via turbulent mixing the traces gas distribution is influenced by the turbulence. This influence is present in the trace gases for a certain time after the turbulence occurrence.

Changes in manuscript: Despite only weak turbulence during the stratospheric leg, the cross isentropic gradient and the related composition change on isentropic surfaces from upstream to downstream the mountain unambiguously conserves the effect of turbulent mixing by gravity wave activity ~~before~~ on the trace gas distribution prior the measurements.

Comment Page 2 Line 1: "Orographic gravity waves [...] may affect the large scale stratospheric circulation." Clearly, there is still a lot to be learned about orographic GW forcing and the effect they have on the general circulation, but is there really any doubt whether orographic GWs have an effect at all?

Reply to comment: True. Changed it accordingly.

Changes in manuscript: Orographic gravity waves play an important role for the thermal and dynamical structure of the atmosphere and ~~may~~ affect the large scale stratospheric circulation

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Comment Page 7 Line 10: What exactly is meant by "analysed PV"?

Reply to comment: We clarified it.

Changes in manuscript: The tropopause was crossed around 18:40 UTC, as indicated by the sharp decrease of the N_2O mixing ratio and the ~~analysed~~ PV interpolated along the flight path.

Comment Figure 4: The wave packet seen between 170.1° E and 170.6° E in the higher altitude leg has a very nice and regular vertical wind w and θ relationship (π / 2 phase shift), just as one would expect from linear wave theory. The same longitude range of the lower altitude leg, however, has an interesting θ structure that does not correspond that well to w. It might be interesting to see if ECMWF IFS predicts similar structures, as these may be related to wave breaking/reflection.

Reply to comment: The large scale structure similarly exists in ECMWF IFS, but the small scale fluctuations are missing because the model resolution is too coarse. For this flight no wave breaking was predicted by the models.

Comment Page 9 Line 1: The word "where" should be replaced with "were".

Reply to comment: True. We changed it accordingly.

Changes in manuscript: The flight sections of the two southern legs ~~where~~ were strongly affected by orographic waves

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Comment Page 9 Line 2: Most literature (and the rest of this paper), provide amplitudes as positive numbers, "+/-" should therefore be omitted for consistency. Also, since fluctuations of potential temperature are mentioned, it would be good to provide their amplitude as well.

Reply to comment: Correct. We changed it accordingly.

Changes in manuscript: Both legs show strong fluctuations of the vertical wind component w and the potential temperature Θ with amplitudes of ~~±2 ms^{-1}~~ 2.5 ms^{-1} and 4.5 K, respectively.

Comment Page 9 Line 7: The phrase "[...] indicative for at least a kinematic breakdown [...]" should probably be replaced with "[...] indicative of at least a kinematic breakdown [...]". Also, the whole sentence is confusing, it is not quite clear what the word "but" in L8 refers to.

Reply to comment: We clarified the sentence and replaced to "but".

Changes in manuscript: At the upper level such ~~a breakdown of scales~~ turbulence is not prominent, although the fluctuations of Θ, w and N_2O (Fig. 3 *(former Fig. 4)*) are indicative ~~for~~ of at least a potential kinematic flux of N_2O, ~~but~~ with only weakly pronounced small scale variability of w'.

Comment Pages 10-11: I may have missed something simple or misinterpreted the terms used, but the discussion of observed GWs in Section 3.3 appears to contain contradictory statements. For example, the terms "lower/upper flight leg" seem to refer to lower/higher altitude flight segments in most of the discussion on P10 and P11. Also, the long horizontal wavelength wave mode is stated to be "totally absent in the lower leg" (P10 L4), and "partly seen in VH in Fig. 3 around 17:45" (P10 L3). However, according to Fig. 3, the aircraft was flying at the lower of the two main altitude levels (i.e. flying the "lower leg"?) around 17:45 UTC. Perhaps in some cases "lower/upper leg" refers to lower/higher altitude, and in some cases to downstream/upstream? In any case, I feel that the terms used for flight segment identification should be updated in the entire Section 3.3, so that no guesswork is needed. For example, one might consider only using the term "flight leg" for a straight (geodesic) flight segments, and adopting other terms to refer to longer portions of the flight.

Reply to comment: Thank you for your thorough reading. On (P10 L7) there was a confusion which we corrected. We also checked the text to make sure that flight leg always denotes a straight, horizontal part of the flight path. The upper flight leg is at 10.9 km and lower flight leg at 7.9 km. Both flight legs include a downstream, an upstream and an above mountain section.

Changes in manuscript: The other, shorter mode in the horizontal wind spectra is well-developed only in the ~~upper~~ lower leg.

Comment Page 11 Line 3: Dissipation is indeed a likely explanation for the change in vertical wave energy flux, but one must not forget that GWs are often reflected at the tropopause, which complicates the interpretation of energy fluxes in this region. In any case, the turbulence observations in this paper provide a stronger argument that wave energy is indeed dissipated in the altitude range considered here.

Reply to comment: That's right. At least, the analysis of the ECMWF model shows no indication wave reflection during the flight (see also Fig. 1b *(former Fig. 2b)*).

Comment Caption of Figure 6: Duplication of the article "the".

Reply to comment: True. We removed it accordingly.

Changes in manuscript: The lower leg lies entirely in the troposphere as indicated by the ~~the~~ dark ~~gray~~ orange data points of N_2O = 328 ppbv.

Comment Page 12 Line 14: Strictly speaking, there is nothing "between Θ < 328.1 K and Θ > 326.3 K", as the two intervals overlap. I would either write "layer between $\Theta = 328.1$ K and $\Theta = 326.3$ K" or, preferably, "a layer with $326.3 \text{ K} < \Theta < 328.1$ K".

Reply to comment: Changed as suggested.

Comment Page 12 Line 15: I cannot see any green crosses in Fig. 6, perhaps this should refer to green squares?

Reply to comment: Correct. We changed it accordingly.

Changes in manuscript: ~~They~~ The intermediate points thus mark a layer between ~~$\Theta < 328.1$ K and $\Theta > 326.3$ K~~ $326.3 \text{ K} < \Theta < 328.1$ K, where the tracer-tracer diagram indicates mixing between the two branches (green ~~crosses~~ squares in Fig. 5 *(former Fig. 6)*)

Comment Page 12 Line 5: The notation "∂N2O / $\partial\theta$" as given in L4 is clear, concise and well-defined. Therefore, I cannot see any benefit of subsequently introducing so many different terms (N2O-gradient, N2O-θ gradient, N2O-θ slope, decrease of N2O with potential temperature θ, N2O decrease with respect to θ, ...) to refer to essentially the same thing.

Reply to comment: We refer to the local idealized N_2O-Θ profile as "$\partial N_2O/\partial\Theta$" (or vertical N_2O-gradient with respect to Θ), when speaking about general aspects of the profile, likewise the term N_2O-Θ relationship, when talking about general aspects of their relation (not necessarily the profile). We further changed consistently to "Θ'/N_2O' ratio" when analyzing the ratio of anomalies relative to the mean and will term this as ratio in general. In some cases we though it is helpful to emphasize certain aspects of this relation related to our hypothesis of tracer changes on isentropes.

Comment Page 14 Line 10: Firstly, notation "θ' N2O$'$ ratio" is a bit odd. "θ / N2O$'$ ratio" or "the ratio of θ' and N2O$'$" would already be better. Secondly, as explained in Section 4.1, the ratio N2O$'$ / θ' depends on integration time and is therefore mathematically not the same as "N2O-θ gradient". The fact that the measured gradients do not actually depend on the integration time too much (in a reasonable range of integration times) is a meaningful finding that supports the main claims of the paper. It is hence important not to confuse the readers by implying these two quantities are one and the same.

Reply to comment: We thank the reviewer for this comment and kept the "Θ' / N_2O' ratio" as stated above and removed the term gradient.

Changes in manuscript: Thus, in case of gravity wave induced turbulent mixing during flight FF09, we expect a ~~steeper vertical N_2O-Θ gradient in the inflow region upwind the mountains (i.e. a smaller Θ' / N_2O' ratio) than at the downstream side of the mountain ridge~~ more rapid decrease of N_2O with increasing Θ in the inflow region upwind the mountains than at the downstream side of the mountain ridge as an effect of turbulent mixing.

Comment Page 18 Line 3: After inspection of Figure 7, it would seem that both θ and N2O concentration amplitudes given here are peak-to-peak values, actual amplitude values should be half that.

Reply to comment: True. Changed it accordingly.

Changes in manuscript: The observed Θ~~-amplitude~~ peak-to-peak variability of 8 K (Fig. 6 *(former Fig. 7)*) would correspond to N_2O = 13 ppbv, while only 4 ppbv are observed consistent with an impact of diabatic mixing processes changing the upstream relation.

Comment Page 18 Line 7: Same issue as P14 L10. I cannot see a good reason for using primed quantities here and unprimed quantities in L9.

Reply to comment: We changed the expressions consistently to the above stated convention.

Changes in manuscript: The downstream impact is ~~This~~ is evident from the different Θ'/N_2O' ~~slope~~ ratio at larger wave lengths at the ~~lee~~ downwind side compared to the upstream ~~slope~~ ratio. The transition between the upstream and downstream ratios occurs at scales <3 km above the mountains. Therefore we conclude that during FF09 mountain waves modified the ~~slope N_2O-Θ~~ Θ'/N_2O' ratio at small scales ~~and induced cross-isentropic turbulent mixing~~. . They induced cross-isentropic turbulent mixing leading finally to changes at large scales downwind the Alps as evident from the ratio Θ'/N_2O' and finally the vertical gradient $\partial N_2O/\partial\Theta$ (Fig. 6 *(former Fig. 7)*).

Comment Caption of Figure 12: Main text discusses various wave time scales, and not length scales. Therefore, the figure should have wave period labels.

Reply to comment: To make it comparable to the other figures we changed the values in the text to wavelength.

Comment Page 18 Line 14: Perhaps the authors meant "negative vertical N2O gradient"?

Reply to comment: Correct. We rearranged the sentence.

Changes in manuscript: Further, the phase relation ~~is constant at 180°, which one would expect for a decreasing vertical N_2O gradient in the stratosphere, but increasing Θ.~~ between the time series of N_2O and Θ (see Fig. 6 *(former Fig. 7)*) is almost constant at 180° for scales < 20 km, which one would expect for opposing vertical gradients of N_2O and Θ in the stratosphere.

Comment Page 20 Line 9: The phrase "periods ranging from 8-16 km" should be replaced with "wavelengths of 8-16 km"

Reply to comment: Changed as suggested.

Changes in manuscript: The last region shows mainly positive trace-gas fluxes with values up to 0.50 ppbv m s^{-1} at ~~periods~~ wavelengths ranging from 8-16 km corresponding to the vertical wind energy maximum around $\lambda_x = 10$ km

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Comment Page 23 Line 30: Again, the expression "The tracer conserves the effect of [...]" should perhaps be rephrased.

Reply to comment: We rephrased the sentence.

Changes in manuscript: The tracer distribution conserves the effect of prior occurrence of ~~the~~ highly transient turbulence occurrence.