

# Response to Reviewers

## GENERAL REMARKS

We thank all three reviewers for their insightful reviews and helpful and constructive comments. Responding to their comments helped improve the manuscript significantly.

All reviewers raised the point that the discussion of the different tropopauses (thermal, dynamical) lacks clarity and is confusing, and also partially misleading. As a general response to all reviewers, we put now the entire discussion of the observations and their interpretation into the framework of the extratropical transition layer (ExTL) and the vertical tracer profiles observed in that region. As discussed e.g. by Hoor et al. (2004), Pan et al. (2010), and Gettelman et al. (2011), it is found that on average the dynamical tropopause is situated slightly below the thermal tropopause and trace gas gradients are more sharp above the thermal tropopause compared to the dynamical tropopause. This is exactly the same behaviour we observe here and the interpretation of our results is now linked to that known feature of tracer characteristics in the ExTL. So, we modified the following sections considerably:

### 1. Introduction:

We added the following paragraphs to the introduction:

“The extratropical upper troposphere and lowermost stratosphere (Ex-UTLS) is characterised by thermal gradients and dynamical barriers which inhibit mixing, give rise to specific trace gas distributions and lead to a variety of definitions of the tropopause (Gettelman et al., 2011; Ivanova, 2013). The thermal tropopause according to WMO criteria (WMO, 1957) is defined as the level, where the lapse-rate decreases to  $2 \text{ K km}^{-1}$  or less and remains so small at least in the overlying layer of 2 km. This definition identifies the vertical change in the static stability and allows for the existence of multiple tropopause layers. The dynamical tropopause is based on the potential vorticity (PV) and includes both changes in static stability and vorticity (i.e., horizontal and vertical wind shear), also viewed as the dynamic stability. The PV values in the stratosphere exceed its values in the troposphere by an order of magnitude. The threshold value of 2 PVU ( $1 \text{ PVU} = 10^{-6} \text{ m}^2 \text{ K s}^{-1} \text{ kg}^{-1}$ ) for separating tropospheric and stratospheric air masses is commonly used in studies on stratosphere–troposphere transport. The chemical tropopause is based on the chemical change at the tropopause, identified from tracer-tracer correlations (Zahn and Brenninkmeijer, 2003), with a threshold value of  $\text{O}_3 \text{ VMR} > 120 \text{ ppbv}$  being used to distinguish stratospheric from tropospheric air (Thouret et al., 2006). The coexistence of different definitions of the tropopause and the observation that characteristics of air masses around the tropopause depend on the applied definition motivated the concept of the extratropical transition layer (ExTL) which describes the extratropical layer around the tropopause; see Gettelman et al. (2011) and references therein.

The vertical distribution of trace species in the Ex-UTLS is controlled by the strong static stability gradients and dynamic barriers to transport in this atmospheric layer. In the case of water vapour, the  $\text{H}_2\text{O}$  VMR is also determined by the coldest temperature the air parcel has experienced on its way to the tropopause (Lagrangian dry/cold point), which decouples the abundance of water vapour from local cross-tropopause mixing to some extent (Hoor et al., 2010; Zahn et al., 2014). The distribution is described by a steep decrease of the  $\text{H}_2\text{O}$  VMR up to the tropopause layer. Across the tropopause layer,  $\text{H}_2\text{O}$  VMR decreases further but less steep until it reaches its near-constant stratospheric value at about 2 km altitude above. The thermal tropopause forms thus an efficient barrier for the large-scale vertical transport of  $\text{H}_2\text{O}$  into the stratosphere, whereas troposphere-stratosphere transport occurs for specific local-scale dynamic situations such as, e.g., tropopause folds (Hoor et al., 2004; Hoor et al., 2010; Gettelman et al., 2011).”

## 2.1 Data coverage and vertical distribution

The discussion of the different definitions of the thermal and dynamical tropopause (lines 147 to 160 of the ACPD paper) was moved to the introduction section for clarity and to avoid duplication. The section on data coverage and vertical distribution contains now a description how the pressure levels for both thermal and dynamical tropopauses were determined and linked to the data set:

“The pressure levels of the thermal tropopause ( $p_{\text{therm.TPH}}$ ) and the dynamical 2 PVU tropopause ( $p_{\text{dyn.TPH}}$ ) were derived from ERA-Interim (Dee et al., 2011) which uses 60 model layers with the top of the atmosphere located at 0.1 hPa. For our analysis, the 6-hourly outputs from ERA-I ( $0:75^\circ \times 0:75^\circ$ ) were interpolated onto a  $1^\circ \times 1^\circ$  horizontal grid and on 60 vertical levels of constant pressure and potential temperature (Kunz et al., 2014; Berkes et al., 2017). Additionally, the variables of the PV, and the pressure of the thermal tropopause ( $p_{\text{therm.TPH}}$ ) based on the WMO criteria were calculated (WMO, 1957; Reichler et al., 2003). The ERA-Interim data were then linearly interpolated with respect to longitude, latitude, pressure, and time onto each flight track with 4 s resolution, as described by Kunz et al. (2014). Interpolated tropopause pressure levels were finally used to determine the position of the aircraft relative to the thermal tropopause or to the 2 PVU iso-surface, respectively, and thus to distinguish whether the aircraft sampled air masses of UT, TPL or LMS origin with respect to the chosen tropopause definition.”

The paragraph on the definition of the seven pressure layers around the tropopause used in our analyses was rearranged for the sake of clarity. It reads now:

“In order to reach both a sufficiently large data set for robust statistical analyses and good vertical resolution, the Ex-UTLS is subdivided into seven layers of 30 hPa thickness each, with three layers located below the thermal tropopause height and three layers above. Thouret et al. (2006) used a similar definition, but referenced to the dynamical tropopause at 2 PVU, i.e. they defined the tropopause as a mixing zone 30 hPa thick across the 2 PVU potential vorticity surface.

The seven layers of 30 hPa thickness each are centred at  $p_{\text{therm.TPH}} = 0$  hPa for the tropopause layer (TPL) itself and then at  $p_{\text{therm.TPH}} \pm 30$  hPa,  $p_{\text{therm.TPH}} \pm 60$  hPa, and finally at  $p_{\text{therm.TPH}} \pm 90$  hPa. From this vertical spacing, the separation of air masses is achieved by applying the following criteria (formulated for the thermal tropopause only):

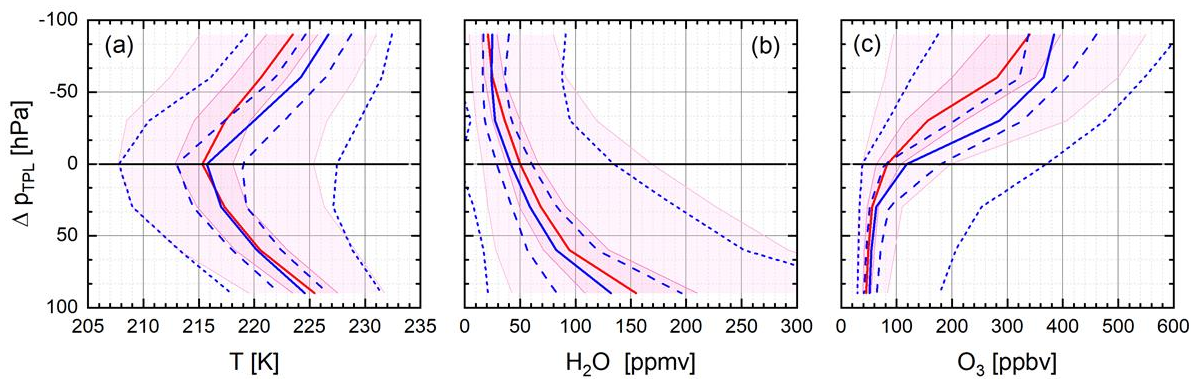
LMS :  $p < p_{\text{therm.TPH}} - 15\text{hPa}$ ; which is limited by the maximum cruise altitude with  $p \approx 190$  hPa;  
TPL :  $p = p_{\text{therm.TPH}} \pm 15\text{hPa}$ ;  
UT :  $p > p_{\text{therm.TPH}} + 15\text{hPa}$ ; limited to lower altitudes by  $p < 350\text{hPa}$ .”

## 3.3 Physico-chemical signature of ice-supersaturated regions in the vicinity of the tropopause

This section is significantly modified. In general terms, the introducing paragraph to this section reads now as follows:

“In order to study the formation history of ISSR and involved processes, we analysed the occurrence frequency and physico-chemical signature of ISSR around the tropopause layer and referred our analyses to both the thermal and the dynamical tropopause. We want to recall the tropopause definitions given in Section 2.1. The thermal tropopause according to WMO criteria (WMO, 1957) is usually seen as an effective transport barrier hampering troposphere-stratosphere exchange, whereas the dynamical tropopause is commonly used for separating tropospheric and stratospheric air masses in studies on stratosphere–troposphere transport since it represents the lower bound of the ExTL. These complementary views on the tropopause have been developed from extensive CO - O<sub>3</sub> analyses, which showed that the 2 PVU surface approximately separates the troposphere from the stratosphere with the ExTL as a transition layer of about 2 km thickness above it and centred on the thermal tropopause (Hoor et al., 2004; Pan et al., 2010; Gettelman et al., 2011). These tracer studies in the extratropics showed that on average the dynamical tropopause is situated slightly

below the thermal tropopause and the gradients of CO and O<sub>3</sub> are much sharper across the thermal tropopause compared to the dynamical tropopause (Hoor et al., 2004; Pan et al., 2010). Similar features are observed for the gradients of temperature T, H<sub>2</sub>O VMR and O<sub>3</sub> VMR, shown in Figure 9 for the North Atlantic region. Similar to the tracer gradients, also the temperature gradient is sharper across the thermal tropopause compared to the dynamical tropopause. In addition, the results confirm the good agreement between the ERA-Interim thermal tropopause height indicated by  $\Delta p_{\text{TPH}} = 0$  hPa (blue lines), the lowest temperatures detected at  $\Delta p_{\text{TPH}} = 0$  hPa, and the chemical tropopause, indicated by O<sub>3</sub> VMR = 120 ppbv at  $\Delta p_{\text{TPH}} = 0$  hPa, and thus the consistency of the used data set. Furthermore, the analysis of the pressure difference between the thermal and dynamical tropopauses reveal an offset of approx. 25 hPa (15 - 35 hPa) which translates into an altitude difference of approx. 1 km (Neis, 2017).



**Figure 9.** Vertical distribution of temperature T (a), H<sub>2</sub>O VMR (b), and ozone VMR (c) relative to the 2 PVU dynamical tropopause and to the thermal tropopause; vertical distributions relative to the thermal tropopause are presented as percentiles [1, 25, 50, 75, and 99] by blue lines and relative to the 2 PVU tropopause conditions by red-shaded areas. “

The concluding paragraph reads now as follows:

“Recalling the structure of the ExTL with the 2 PVU dynamical tropopause at its lower bound separating the stratosphere from the troposphere, and centred on the thermal tropopause, we find that on the top of the ExTL non-ISSR air masses show a clear stratospheric signature, while ISSR air masses are still strongly influenced by mixing and carry a significant tropospheric fingerprint compared to the non-ISSR air masses. Above the dynamical tropopause and thus inside the ExTL, the influence of mixing increases gradually for both ISSR and non-ISSR air masses and the difference in troposphericity is much less pronounced than near the top of the ExTL.”

Detailed responses to reviewers concerns are given in the specific responses.

## Response to Reviewer #1

**Reviewer:** *This manuscript uses a humidity data set from commercial aircraft to analyze humidity and ice supersaturated regions in the upper troposphere. The manuscript is quite good, with careful and comprehensive analysis of the uncertainties and corrections in the data set. However, some of the analysis doesn't quite make sense, especially the discussion of thin cloud occurrence and correspondence of sign with NAO at the end of the manuscript. That analysis needs significant modification as I outline in the specific comments below. Except for this and minor comments, this manuscript should be publishable in ACP with appropriate revisions.*

**Authors:** We appreciate the valuable review and respond in detail to the comments in the following.

### MAJOR COMMENTS

#### **Tropopause definitions and roles:**

**Reviewer:** *Page 14, L413: this bothers me a bit. The thermal tropopause is a robust barrier in the tropics, but here the average RH is 80% or less at the tropopause, so it does not need to be a robust barrier. Also motion is not purely vertical here, but more horizontal and isentropic. Please explain.*

**Authors:** In contrast to larger-scale horizontal and isentropic transport processes which dominate troposphere-stratosphere exchange, we focus here on local vertical transport pathways, since these processes may influence the formation of ISSR within and above the tropopause layer. In addition we put now the entire discussion of the observations and their interpretation into the framework of the extratropical transition layer and the vertical tracer profiles observed in that region. As discussed e.g. by Hoor et al. (2004), Pan et al. (2010), and Gettelman et al. (2011), it is found that on average the dynamical tropopause is situated slightly below the thermal tropopause and trace gas gradients are more sharp above the thermal tropopause compared to the dynamical tropopause. This is exactly the same behaviour we observe here. To support this finding we added the vertically resolved data for O<sub>3</sub> VMR and H<sub>2</sub>O VMR with respect to the thermal and dynamical tropopauses as new Figure 9.

In summary, we modified the Introduction and Section 3.3 considerably; see the general remarks at the top of this document.

**Reviewer:** *Page 16, L480: why is the thermal tropopause a transport barrier and the dynamical tropopause in extratropics not a barrier? I'm not sure I understand your logic here.*

**Authors:** As explained above, we put now the entire discussion of the observations and their interpretation into the framework of the extratropical transition layer. The last paragraph of Section 3.3 summarising the results, was removed.

#### **Cirrus clouds and ice-supersaturation:**

**Reviewer:** *Page 18, L545: I don't think the casual analysis of the frequency of cirrus is helpful. Cirrus frequency is a function of instrument as you note. And cirrus layers need not be supersaturated, so there need be no link here.*

**Reviewer:** *Page 18, L548: I do not think you can argue that just because cirrus and ISSRs have about the same frequency (But ISSR is lower from the best and most sensitive sensor), that most cirrus occur in ISSRs. You need to show physical and temporal coincidence.*

**Authors:** We agree that the results of our comparison of ISSR occurrence from MOZAIC and of cirrus cloud occurrence from satellites might appear over-interpreted. Nevertheless, we believe that this comparison is useful for several reasons. First, there is a physical link between the formation of cirrus clouds and ice-supersaturation, since ice crystal formation only takes place at high ice-supersaturation. In addition, ice-supersaturation may occur inside cirrus also during their lifecycle, e.g., when the cloud is further lifted up while sedimentation leads to the reduction of available ice crystals for the further deposition of water vapour; see e.g. Krämer et al. (2016). Second, the lateral resolution of observations is significantly different between satellites and MOZAIC in-situ data, so that local-scale in-cloud fluctuations in RH<sub>ice</sub> might be seen by MOZAIC but not by the satellite

instrument. And finally, our motivation for this pilot comparison exercise was to investigate whether the observation probabilities of ISSR in MOZAIC and cirrus cloud occurrence in satellite data is of the same order of magnitude. For sure, our pilot exercise cannot replace a detailed study that picks up your valuable arguments, but will initiate this work. For these reasons we decided to keep this section in the manuscript. However, we softened our conclusions considerably, reflecting your concerns.

In response to your arguments, we added the following paragraph to the introduction to Section 3.4:

“Furthermore, the analysis of a large set of combined observation of  $RH_{ice}$  and ice crystal number concentration  $N_{ice}$  during a series of research flights (approx. 68000 observations of ice-supersaturation; Krämer et al., 2016) demonstrated, that approx. 80 % of the observed ice-supersaturation events are associated with in-cloud conditions. On the other hand,  $RH_{ice}$  probability distribution functions inside cirrus clouds are characterised by most probable values at or slightly above ice-saturation at  $RH_{ice} = 100\%$  (Krämer et al., 2009; Diao et al., 2014; Diao et al., 2015; Petzold et al., 2017) which means that cirrus clouds exist to a considerable fraction also in ice-subsaturated air masses, depending on their state of life.”

The concluding paragraph of Section 3.4 was significantly softened and reads now:

“The good agreement between MOZAIC in-situ observations of ISSR occurrence with the high-cloud fraction from satellite instruments encourages further detailed studies on this matter. First exemplary analyses of simultaneous observations of  $RH_{ice}$  and  $N_{ice}$  which are now possible within the ongoing IAGOS programme already indicate a strong correlation of high  $RH_{ice}$  values with its occurrence inside cirrus clouds (Petzold et al., 2017). “

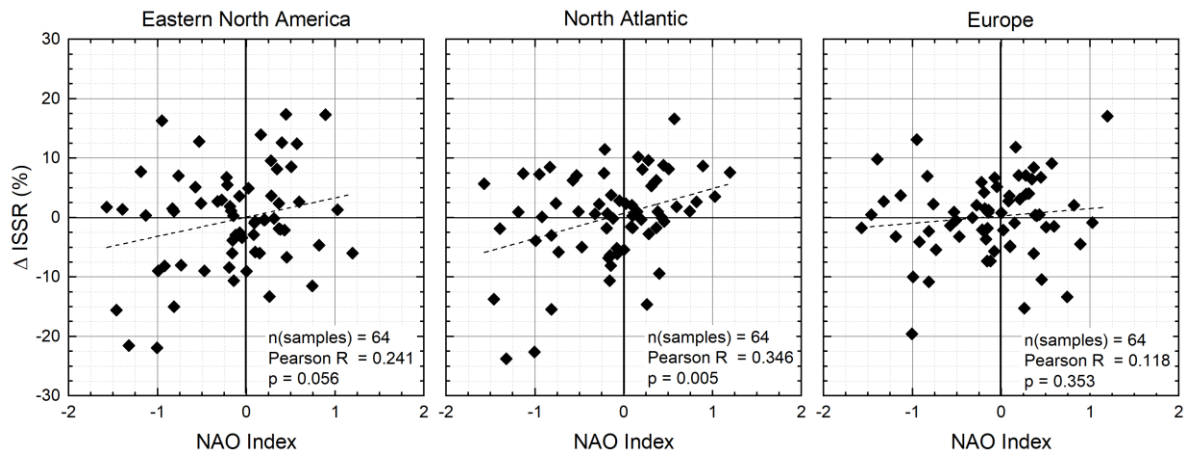
#### **ISSR and NAO index:**

**Reviewer:** Page 21, L607: *I’m not sure I follow this here. If there is a non-zero correlation then there is a correspondence of signs is there not? Not familiar with the method.*

**Reviewer:** Page 21, L613: *why does the same sign in a bit over half the cases mean statistical significance? Isn’t 50% totally random?*

**Authors:** The intention of this analysis was to investigate a potential link between the deviation of ISSR occurrence from the long-term average on one hand and the NAO index as an indicator of storm track activity on the other hand. As is obvious from the distribution of the data pairs in Figure 15, the correlation is weak in all cases. Equal occurrence of cases with positive correlation of signs, i.e., positive NAO index associated with positive deviation of ISSR occurrence from the long-term average and vice versa, and the occurrence of cases with negative correlation of signs, i.e., positive NAO index associated with negative deviation of ISSR occurrence from the long-term average and vice versa, indicates random occurrence. This is certainly the case for the region of Eastern North America (Fig. 15, left panel). Particularly for the region North Atlantic, the positively correlated cases occur at 61% of the observations and the negatively correlated cases at 39%. Over the Europe, this difference is again weaker.

To make our analysis of this potential link more robust, we skipped now the distinction between positively and negatively correlated signs and performed a cross-correlation analysis. The results of this analysis are now shown in the revised Figure 15 (now 16):



**Figure 16.** Correlation analysis with respect to the correlation of signs between NAO index and deviation of ISSR occurrence from the long-term average ( $\Delta$  ISSR) for the target regions; numbers indicate the results from the correlation analysis with respect to number of samples  $n$ , Pearson  $R$  and significance level  $p$ .

The values of the correlation coefficient  $R$  and the significance level  $p$  indicate that for the North Atlantic this correlation is significant at a level of 99%. For the two other regions the correlation is not significant.

The description of the figure and the conclusions drawn read now:

“For the regions Eastern North America and Europe the correlation between NAO index and  $\Delta$  ISSR fraction is not statistically significant. For the North Atlantic however, the results of the cross-correlation analysis indicate statistical significance at a level of 99%. The obtained correlation of signs is in line with the observation that the occurrence of ice-supersaturation is well correlated with the storm track activity (Spichtinger et al., 2003b; Gettelman et al., 2006; Lamquin et al., 2012).”

We also rephrased the respective statement in the Conclusions section, which reads now:

“Yet, we identify a significant correlation of signs between the NAO index and the deviation of seasonal ISSR occurrence probabilities from the long-term average for the North Atlantic, whereas no such correlation was found for Eastern North America and Europe.”

**Reviewer:** Page 21, L615: How does a correspondence of anomalies correlate with the storm track?

**Authors:** To motivate the expected link between storm track activity and higher ISSR occurrence we include the following sentence in the introduction to this part of our analysis:

“As an example, a positive value of the NAO index indicates that  $\Delta p$  (Iceland L to Azores H) is larger than on average. This larger pressure difference causes stronger westerly winds and thereby more active storm tracks over the North Atlantic. Under such conditions we would expect a higher probability of ice-supersaturation in the uppermost troposphere due to more frequent warm conveyor belts that can induce the formation of ISSRs in the upper troposphere (Spichtinger et al., 2005).”

## SPECIFIC COMMENTS ABOUT THE DATA SET AND THE DATA ANALYSIS PROCEDURE

**Reviewer:** *Page 4, L127: what is the horizontal resolution?*

**Authors:** We added the requested information and rephrased the sentence as follows:

The horizontal resolution of our data set is 1 km, set by the instrument time resolution of 4 s and the cruising speed of approx. 250 m s<sup>-1</sup>. The vertical resolution is set to 30 hPa, which corresponds to a vertical distance of approx. 750 m at cruise altitude (Thouret et al., 2006) and assures sufficient statistical robustness of the conducted analyses. This vertical resolution is of similar order as the typical resolution of UTLS data with a vertical grid spacing of about 50 hPa in the vicinity of the tropopause (Reichler et al., 2003).

**Reviewer:** *Page 4, L141: what does 30 - 65 hours mean? In each grid box? How many flights per grid box per season?*

**Authors:** The descriptions means that per regional box between 30 hours and 65 hours of flight per season (3 months) were collected, which corresponds to 27,000 to 60,000 data points of 4 s duration each, per season per year. We rephrased the description as follows:

“The annual data coverage for each analysed regional box varies between 30 and 65 flight hours of MOZAIC aircraft per season (3 months) which corresponds to 27,000 to 60,000 data points of 4 s duration each, per season per year.

**Reviewer:** *Page 5, L166: what is the vertical and horizontal resolution of ERA-I in the UTLS?*

**Authors:** We rephrased the sentence for clarification as follows:

“The pressure levels of the thermal tropopause ( $p_{\text{TPHWMO}}$ ) and the dynamical 2 PVU tropopause ( $p_{\text{TPHDYN}}$ ) are derived from ERA-Interim (Dee et al., 2011) which uses 60 model layers with the top of the atmosphere located at 0.1 hPa.”

**Reviewer:** *Page 5, L178: wouldn't it be better to have PDFs in each season since it seems there are a lot of data points. More statistics than the mean it seems are available.*

**Authors:** This is exactly what we did. In order to clarify our procedure, we rephrased the description as follows:

“Since each data set from one single flight provides only a one-dimensional snapshot of the state of the atmosphere along the flight track, and each aircraft cruises at a slightly different pressure level, the entire MOZAIC data are consolidated to season files of 3-months season files duration, allowing the analysis of vertical distributions of atmospheric state parameters on a robust statistical basis. For each season file, the statistical distribution (average and standard deviation, percentiles) of investigated properties (temperature, O<sub>3</sub> VMR, H<sub>2</sub>O VMR, RH<sub>ice</sub>, ISSR fraction) is calculated with respect to the above defined UT, TP and LMS vertical layers. From these seasonal averages or percentiles, respective 15-year values including their variability are determined.”

**Reviewer:** *Page 8, L270: was Research flight data selected for the same geographic regions as MOZAIC data shown in Figure 5?*

**Authors:** The MOZAIC data shown in Figs. 3 -5 refer to the complete data set. i.e., data include also measurements outside the geographic regions our analysis is focusing on. In consequence, the research aircraft data were also not restricted to regions but only to temperature and pressure ranges.

To clarify this, we rephrased the first paragraph of Section 2.4:

“The IFC method was applied to the full reanalysis data set from 1995 to 2010. Figure 3a illustrates the effect of the IFC method for the averaged RH<sub>ice</sub> PDF for the entire MOZAIC data set, irrespective of the geographical regions the data were collected. The presented average PDF and variability is calculated from annual PDFs.”

**Reviewer:** *Page 11, L353: Table 1 just restates the right column from Figure 8 correct? Maybe it is not necessary? Can you put the standard deviations from Figure 2 on the plot in Figure 8?*

**Authors:** Indeed, Table 1 restates the right column from Figure 8. However, we also want to give average values and standard deviations in a numerical format for comparison with our studies. We also added standard deviation from Table 2 to Figure 8, but then Figure 8 becomes highly unclear and hard to understand. Finally, our decision was to present only average values in Figure 8 and add numerical values in Tables 1 and 2.

**Reviewer:** *Page 13, L396: not exactly clear to me how this is different than the relevant panel in Figure 8. Just adding the dynamic tropopause?*

**Authors:** We added respective values for the dynamical tropopause, but skipped the separation into seasons. The latter step was mainly made because we wanted to analyse physico-chemical signatures of sub- and supersaturated air masses by analysing additionally the O<sub>3</sub> VMR for these air masses. Since MOZAIC H<sub>2</sub>O and O<sub>3</sub> data sets are not completely overlapping due to instrument performances, we decided to skip the distinction into seasons to generate a larger data ensemble for the targeted statistical analyses.

For clarity, we added the sentence “Please note that the ISSR fraction values compiled for the thermal tropopause correspond to the values listed in Table 2, but without separation into seasons.”

**Reviewer:** *Page 14, L435: where does the ozone come from? Also MOZAIC I assume? Please specify. What is the minimum detectable concentration? And can you provide a validation reference and maybe a sentence or two.*

**Authors:** Indeed, the origin of the O<sub>3</sub> data needs to be introduced. We added the following sentence to Section 2.2 which is also renamed to RH and O<sub>3</sub> instrumentation:

“Since the launch of MOZAIC, the programme provides also O<sub>3</sub> VMR data in addition to H<sub>2</sub>O and RH<sub>ice</sub> observations. Aboard MOZAIC and now IAGOS aircraft, ozone is measured by means of a UV absorption instrument which is characterised by an instrument noise of  $\pm 2$  ppbv and an integration time of 4 s (Nédélec et al., 2015). We used the collocated measurement of O<sub>3</sub> and H<sub>2</sub>O / RH<sub>ice</sub> for the characterisation of ice-supersaturated air masses with respect to a potential stratospheric influence.”

## MINOR CORRECTIONS

**Reviewer:** *Page 2, Line 67: embedded*

**Authors:** replaced as suggested

**Reviewer:** *Page 3, L78: close to the tropopause. Which tropopause? Thermal is specified in the next sentence.*

**Authors:** Sentence rephrased: “In contrast to the strong negative gradient in H<sub>2</sub>O VMR at altitudes below but close to the thermal tropopause, ISSR occur frequently in the humid and cold upper tropospheric air masses.”

**Reviewer:** *Page 3, L82: again, which tropopause definition?*

**Authors:** “Thermal” added.

**Reviewer:** *Page 8, L259: Figure 4. The RH<sub>ice</sub> line looks solid to me as well.*

**Authors:** Figure 4 was adjusted accordingly.

**Reviewer:** *Page 10, L308: might be better to state that specific humidity is lower in summer over E. N. America in the UT.*

**Authors:** We agree and rephrased the sentence as:

“...while for the Eastern North American region the upper free troposphere layers seem to exhibit higher specific humidity be more humid in winter than respective air masses over the ocean.”



**Reviewer:** *Page 11, L332: the vote part of our study focuses on the vertical....*

**Authors:** For the sake of clarity we rephrased the sentence as: "Our study is focusing on ..."

**Reviewer:** *Page 15, L449: please define 'their' with a reference. Assume it is the same as previous paragraph, but please be specific.*

**Authors:** We rephrased the sentence to be more specific:

"Using the O<sub>3</sub> VMR as a stratospheric air mass tracer and adapting the approach of Cirisan et al. (2013), we define the troposphericity parameter  $m$  for an ensemble of data characterised by median (med) and 99 percentile (P99) values as ..."

**Reviewer:** *Page 18, L531: extra space in years*

**Authors:** Typo was corrected.

## Response to Reviewer #2

**Reviewer:** *This paper presents an analysis of data of relative humidity for the period 1995 to 2010, obtained via instrumented passenger aircraft in the framework of IAGOS and MOZAIC over the northern mid-latitudes (40-60N) in 3 longitude ranges: Northeast America, North Atlantic, and Europe. The huge amount of data makes it possible to cover several vertical altitude ranges of 30 hPa thickness with sufficient data density to allow robust statistics. The altitude bands are defined with respect to the thermal and the dynamical tropopause, respectively, and the "troposphericity" (i.e. the fraction of tropospheric origin in an air parcel) is determined using simultaneous data of ozone VMR. The focus of the study is ice supersaturation.*

*The data show, that ISSRs (ice supersaturated regions) occur most often directly below the thermal tropopause, rarely directly above it, and almost never further up in the stratosphere. There is a distinct seasonal cycle in all 3 considered regions, but no significant trend over the 15 years of the study. The North-Atlantic Oscillation seems to have an influence on the occurrence of ISSR over the North Atlantic and Europe, but not over North America, which is physically plausible. ISSRs are colder and moister than their subsaturated surroundings (in agreement with earlier results), and they are poorer in ozone and have accordingly a larger troposphericity than the subsaturated environments, which is plausible as well considering the fact that most ice supersaturation is formed by uplifting of airmasses. The data show also that ice supersaturation is very closely related to cloudiness, that is, most ice supersaturation is found within clouds.*

*Thus, this paper provides a number of new and interesting results. It is well written for the most part. There are only a few points where I think the presentation can be made clearer and where perhaps the discussion can consider one or two more points. The paper should surely be published after the issues below are addressed.*

**Authors:** We thank the reviewer for the positive feedback and the valuable review and respond in detail to your comments in the following.

### MAJOR COMMENTS

**Reviewer:** *The paragraph lines 388 to 395 should be reworked; it is unclear what you did. For instance, what is an "average occurrence probability"? Do you mean the average frequency of occurrence or something else? What is the pdf of ISSR occurrence? Is this simply the probability of ISSR occurrence? I also do not understand what the distinction between seasons has to do with statistical quantities like median and percentiles and how these two non-related things are linked here in one sentence. And finally, what is the statistical entity?*

**Authors:** We agree that the paragraph is confusing. In fact, "average occurrence probability" refers to the average frequency of occurrence, as supposed by the reviewer. Also, "PDF of ISSR occurrence" refers to the probability of ISSR occurrence. The entire confusing explanation why we focused in this part of our analysis on the entire data set and skipped distinctions into seasons is removed.

Summarising, we rephrased the paragraph:

"Our analysis of ISSR occurrence in the vicinity of the exTL is confined to the North Atlantic region, for which we have the highest data density available with respect to vertical resolution. As described generally in Section 2.1, the entire data set of individual  $RH_{ice}$  observations over the North Atlantic region was divided into yearly subsets for seasons DJF, MAM, JJA, and SON. For each year, season and altitude layer relative to the thermal and dynamical tropopauses, the average frequency of occurrence of observations with  $RH_{ice} > 100\%$  was determined. The probability of ISSR occurrence per altitude layer with respect to the entire period of 15 years was then calculated from this record of seasonally averaged ISSR frequencies of occurrence. The results are compiled in Table 3 for both tropopause definitions used here. Please note that the ISSR fractions compiled for the thermal tropopause correspond to the values listed in Table 2, but without distinction for seasons."

**Reviewer:** *The comparison between statistics relative to the thermal and the dynamic tropopauses is not easy to understand, perhaps because it is unclear what exactly has been done. The first issue that must be clarified is whether the tropopause pressure and the pressure of the 2 PVU surface are available for each single measurement or are there only average values available, which would be bad for the analysis. What is the average  $\Delta p$  with respect to (wrt) the thermal tropopause of the 2 PVU surface? It seems that 2 PVU occurs quite often or in the majority of cases in the UT1 layer. This should be stated. However, it does not seem as if the mean profiles wrt to the dynamical TP are just shifted versions of the profiles wrt the thermal TP definition. Is this a consequence of averaging or why is this so? Next, Table 3 lists under Thermal TP numbers which I expected to be annual mean values of numbers in Table 1 under AVG, but a quick calculation shows something different. Is this because of different weights for the seasons or what is the reason? (For instance take the  $20:0 \pm 6:5$  in column 3 of Table 3. Should this not be the mean of 21:3, 18:6, 17:1 and 18:4 in the right hand box AVG in table 1?). And finally, Fig. 10 shows different behaviour in the left and right panels. Although you give a good physical explanation, I am not fully convinced. In the thermal TP coordinates there is a strong difference between ISSR and non-ISSR already at 30 hPa above the TP, but in the dynamical TP version there is only a much smaller difference at 60 hPa above the 2 PVU level. Is it possible that, on average, the 2 PVU surface is more than 60 hPa below the thermal TP? Eventually we should expect to see qualitatively the same profiles, irrespective of the actual choice of a vertical coordinate, isn't it?*

**Authors:** As explained in the GENERAL REMARKS in response to all reviewers, we put now the entire discussion of the observations and their interpretation into the framework of the extratropical transition layer (ExTL) and the vertical tracer profiles observed in that region. See details hereto in the response to all reviewers.

We will now focus on specific responses to the Reviewer's concerns:

*1. The comparison between statistics relative to the thermal and the dynamic tropopauses is not easy to understand, perhaps because it is unclear what exactly has been done.*

**Authors:** In response to this criticism we introduced a description of the methodology into Section 2.1; see the response to all reviewers above.

*2. It seems that 2 PVU occurs quite often or in the majority of cases in the UT1 layer. This should be stated. However, it does not seem as if the mean profiles wrt to the dynamical TP are just shifted versions of the profiles wrt the thermal TP definition. Is this a consequence of averaging or why is this so?*

**Authors:** The vertical distributions of properties with respect to the two tropopause definitions were determined independently and cannot be simply transferred into each other. The explanation is that the vertical spacing by  $\Delta p = 30$  hPa starts from two different pressure levels  $p_{\text{therm.TPH}}$  and  $p_{\text{dyn.TPH}}$  which are not connected by a straightforward relationship. Over the North Atlantic we found an offset of approx. 25 hPa between dynamical and thermal tropopause with variations from 15 to 35 hPa.

A sentence stating this result was included in section 3.3, reading: "Furthermore, the analysis of the pressure difference between the thermal and dynamical tropopauses reveal an offset of approx. 25 hPa (15 - 35 hPa) which translates into an altitude difference of approx. 1 km (Neis, 2017)."

*3. Table 3 lists under Thermal TP numbers which I expected to be annual mean values of numbers in Table 1 under AVG, but a quick calculation shows something different. Is this because of different weights for the seasons or what is the reason? (For instance take the  $20:0 \pm 6:5$  in column 3 of Table 3. Should this not be the mean of 21:3, 18:6, 17:1 and 18:4 in the right hand box AVG in Table 1?).*

**Authors:** The values listed in Table 3 are calculated from the entire data set and not from seasonally averaged values to get the full variability of observations. However, differences to the mean values calculated from Table 1 seasonal means are small:

The value of  $20.0 \pm 6.5\%$  from column 3 in Table 3 corresponds to the mean of 22.4, 20.1, 20.8 and 20.4 which is 20.9. It has to be noted that only values from Table 1 for the North Atlantic region should be used. The columns of Table 3 named AVG refer to the seasonal means over all three regions. This is clarified by modifying the column description in Table 3 to “AVG(ENA, NATl, EU).

4. Fig. 10 shows different behaviour in the left and right panels. Although you give a good physical explanation, I am not fully convinced. In the thermal TP coordinates there is a strong difference between ISSR and non-ISSR already at 30 hPa above the TP, but in the dynamical TP version there is only a much smaller difference at 60 hPa above the 2 PVU level. Is it possible that, on average, the 2 PVU surface is more than 60 hPa below the thermal TP? Eventually we should expect to see qualitatively the same profiles, irrespective of the actual choice of a vertical coordinate, isn't it?

**Authors:** As explained before, the vertical distributions of properties with respect to the two tropopause definitions were determined independently and cannot be simply transferred into each other. In addition, we found an average pressure difference between the thermal and dynamical tropopauses of approx. 25 hPa (15 - 35 hPa).

As said above, the 2 PVU surface approximately separates the troposphere from the stratosphere with the ExTL as a transition layer of about 2 km thickness above it and centred on the thermal tropopause. Hence, the two tropopauses describe completely different positions within the ExTL so that we cannot expect similar profiles relative to the two tropopause definitions since different processes come into play.

## Minor Issues

**Reviewer:** Occasionally the term UTH is used. This should be avoided. UTH is a radiance based measure of a kind of mean relative humidity in a thick layer in the upper troposphere; it is a non-local measure. In contrast, IAGOS and MOZAIC yield local measures of relative humidity, and even after averaging over certain layers they should not be called UTH to avoid confusion. Better call it "the relative humidity field of the UT" or similar, but avoid UTH.

**Authors:** As recommended, we replaced the term UTH throughout the manuscript by RH or similar.

**Reviewer:** The last sentence of the introduction should be changed. The middle atmosphere is hardly relevant for IAGOS.

**Authors:** This was a typo, we simply removed the reference to the middle atmosphere. The sentence reads now: “Chemistry-climate models like L90MA and L47MA use a vertical grid spacing of 15 - 25 hPa near the extratropical tropopause (Jöckel et al., 2016) which is reflected in the selected vertical resolution of MOZAIC data layers.”

**Reviewer:** Figure caption 1: I do not understand what you mean with the pdf of data points. Do you mean simply the number of measurements or the fraction of measurements in a certain grid box?

**Authors:** We mean the fraction of measurements in a certain grid box. The modified figure caption is: “**Figure 1.** Global coverage of water vapour observations by MOZAIC for the period 1995 to 2010, shown as decadal logarithm of the probability distribution function (PDF) of the data points (fraction of measurements in a certain grid box); red boxes indicate the target areas for our analyses.”

**Reviewer:** Line 251: "Figure 4 illustrates ... of RH...": is this with or without IFC applied?

**Authors:** We specified the data set as “Distribution of  $RH_{ice}$  for the entire MOZAIC period from 1995 to 2010 with IFC applied as a function of ambient temperature with the colour indicating the probability of occurrence ...”

**Reviewer:** *Figure 4: Please describe how these data are normalised. Is the sum over the whole figure 1?*

**Authors:** We specified: "Figure 4 illustrates the distribution of RH<sub>ice</sub> observations from the entire MOZAIC data set shown in Figure 1 as a function of ambient temperature, colour-coded by the probability of occurrence, i.e. the fraction of data points for a specific combination of temperature and RH<sub>ice</sub> with the respect to the entire ensemble."

**Reviewer:** *I am a bit puzzled by the kind of averages applied. In line 176 it says " data are consolidated to 3-months season files", but in line 292 we have monthly mean profiles. Furthermore, is the distance of the current pressure level of a single 4-sec data point to the tropopause pressure recorded for every data point, or is the tropopause pressure averaged over a month and this average taken as reference (which would be a bad strategy to my view)?*

**Reviewer:** *Figures 6 and 7: why do you use geometrical height instead of the  $\Delta p$  for these figures?*

**Authors:** The calculation of monthly mean profiles and the use of km as vertical spacing was done exclusively for producing Figs. 6 and 7 which can be compared directly to respective figures from Zahn et al. (2014). All statistical analyses have been performed using 3-months season files. The pressure distance of the aircraft position to the thermal and dynamical tropopause is determined for each single data point; the procedure is described in Section 2.1.

**Reviewer:** *Line 373/4: The statement may be wrong or perhaps right for the wrong reason. If the mean value of a positive quantity gets small, the variability usually gets smaller as well. Thus I suggest you to consider instead of  $\sigma$  the normalised  $\sigma$ :  $\sigma/\mu$  (i.e. std. deviation divided by mean value).*

**Authors:** Good point! We replaced the standard deviations in Table 2 by the normalised standard deviations and rephrased as follows: "Focussing on the UT layers, the relative standard deviations of the ISSR fractions are highest for the lowest layer investigated here, at least for winter and spring seasons for which the largest ISSR fractions are found."

**Reviewer:** *Comparison with RS Lindenberg (Figure 12a): has the same pressure band be selected for the RS data as for the MOZAIC/IAGOS data or are these the plain overall figures from the old publication?*

**Authors:** We did not reanalyse the Lindenberg data but refer to the original publication.

**Reviewer:** *It is not clear why CALIPSO can have higher cloud frequency than ISSR frequency. The argument that CALIPSO sees subvisible cirrus (SVC) explains only that it sees more than other satellite instruments do, unless SVC can survive in subsaturated air for a quite long time, where it is unclear to me what quite long actually means. I think that the reason for this result is rather in the difference of local vs. non-local measurements, just as the cloud fraction in a single level is always smaller than the cloud coverage over several levels.*

**Authors:** According to Stubenrauch et al. (2010), the high cloud amount (HCA) of CALIPSO is about 10% larger than HCA of CALIPSO for clouds excluding subvisible cirrus. Therefore, the difference can be attributed to instrument sensitivities. In order to explain more in-depth we rephrased the statement about CALIPSO to:

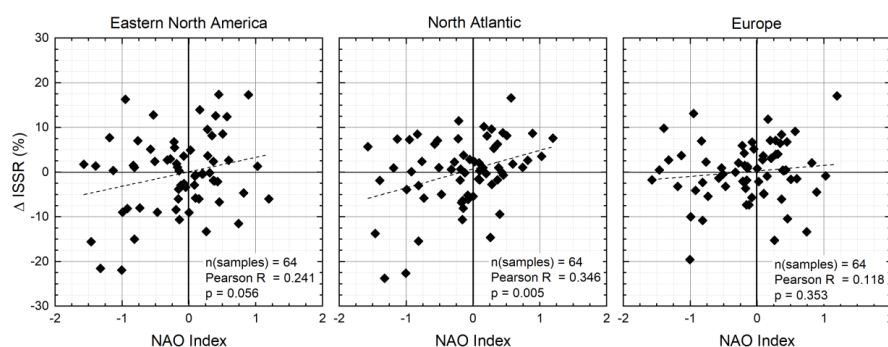
"According to Stubenrauch et al. (2010), the high cloud fraction of CALIPSO is about 10% larger than respective values of CALIPSO for clouds excluding subvisible cirrus. Therefore, the difference between high cloud fractions from CALIPSO and from the other instruments shown in Figure 13 can be attributed to instrument sensitivities."

**Reviewer:** *Final paragraph of 3.5: Misuse of "cross-correlation". A cross-correlation is simply a correlation between two different quantities (as opposed to auto-correlation). Furthermore I suggest to replace "probability" in this paragraph with "fraction" in order to avoid wrong connotations. I am also a bit unhappy with "correlation" since I do not see that the two time-series have been correlated*

(in this case indeed cross-correlated) which would easily be possible. In this case there are also standard techniques to evaluate the statistical significance of the result (i.e. whether the correlation coefficient is significantly different from zero). The sentence "we consider ... statistically significant" should be deleted. This is not a question of "consideration" but of calculation. However, the physical explanation for your result is plausible. In the same sense, the statement in line 645 "significant correlations..." should be reformulated, for instance "physically plausible influence of the NAO on ISSR occurrence is detected in the time series...).

**Authors:** The entire section was modified in refigure 13sponse to a comment by Reviewer #1. We performed a cross-correlation analysis for all three regions and show the results in the modified Figure 16 (formerly 15). This cross-correlation analysis reported a statistically significant correlation between NAO index and deviation of ISSR occurrence from the long term average for the North Atlantic, but no significant correlations for the other two regions. The description of the figure and the conclusions drawn read now:

"For the regions Eastern North America and Europe the correlation between NAO index and  $\Delta$  ISSR fraction is not statistically significant. For the North Atlantic however, the results of the cross-correlation analysis indicate statistical significance at a level of 99%. The obtained correlation of signs is in line with the observation that the occurrence of ice-supersaturation is well correlated with the storm track activity (Spichtinger et al., 2003b; Gettelman et al., 2006; Lamquin et al., 2012)."



**Figure 16.** Correlation analysis with respect to the correlation of signs between NAO index and deviation of ISSR occurrence from the long-term average ( $\Delta$  ISSR) for the target regions; numbers indicate the results from the correlation analysis with respect to number of samples n, Pearson R and significance level p.

**Reviewer:** The data show that "by far the largest part of ISSR occurs inside cirrus clouds". You should ask yourself: Why? Doesn't this imply that most ISSR reach the humidity threshold for heterogeneous or even homogeneous freezing shortly after the air mass began to be supersaturated? Are there further implications? Since the NH has more heterogeneous IN than the SH, do you expect that on the SH a smaller fraction of ISSR is inside clouds?

**Authors:** In response to a comment by Reviewer #1 we added the following paragraph to the introduction to Section 3.4:

"Furthermore, the analysis of a large set of combined observation of  $RH_{ice}$  and ice crystal number concentration  $N_{ice}$  during a series of research flights (approx. 68000 observations of ice-supersaturation; Krämer et al., 2016) demonstrated, that approx. 80 % of the observed ice-supersaturation events are associated with in-cloud conditions. On the other hand,  $RH_{ice}$  probability distribution functions inside cirrus clouds are characterised by most probable values at or slightly above ice-saturation at  $RH_{ice} = 100\%$  (Krämer et al., 2009; Diao et al., 2014; Diao et al., 2015; Petzold

et al., 2017) which means that cirrus clouds exist to a considerable fraction also in ice-subsaturated air masses, depending on their state of life.”

The concluding paragraph of Section 3.4 was significantly softened and reads now:

“The good agreement between MOZAIC in-situ observations of ISSR occurrence with the high-cloud fraction from satellite instruments encourages further detailed studies on this matter. First exemplary analyses of simultaneous observations of  $RH_{ice}$  and  $N_{ice}$  which are now possible within the ongoing IAGOS programme already indicate a strong correlation of high  $RH_{ice}$  values with its occurrence inside cirrus clouds (Petzold et al., 2017). “

**Language, typos, etc.**

**Reviewer:** *Line 71: remove comma after supersaturation.*

**Authors:** Done.

**Reviewer:** *Lines 227/8: Details of ... in detail .. Please reformulate.*

**Authors:** Done, removed ... in detail ...

**Reviewer:** *Line 275: Please replace "validation" with "comparison". And then "The MOZAIC ... IS plotted..."*

**Authors:** Done.

**Reviewer:** *Lines 327 and 329: change to "north" and "south" (i.e. use lower case).*

**Authors:** Done.

**Reviewer:** *Line 340: I suggest to write "Similar AVERAGE values of ..."*

**Authors:** Done.

**Reviewer:** *Line 353: delete "set".*

**Authors:** We replaced the sentence by “...and in the last set of columns averaged over all regions” since we refer here not only to one column but to a block of four columns.

**Reviewer:** *Line 491: add comma after dynamics.*

**Authors:** Done.

**Reviewer:** *Line 579: thus HAS positive ...*

**Authors:** Modified to “...thus show positive ...”

**Reviewer:** *Figure 15: in my printout there is no grey shading.*

**Authors:** Figure has been replaced, see annotated manuscript.

**Reviewer:** *Line 596: warMer and moister (or do you indeed mean more, i.e. a larger quantity of moist air?)*

**Authors:** The phrase “which brings warmer and more moist air to Europe” is removed.

**Reviewer:** *Line 613: MOZAIC (with I).*

**Authors:** Done

### Response to Reviewer #3

**Reviewer:** This manuscript describes in situ measurements of relative humidity (RH) in the upper troposphere and lower stratosphere (UTLS) from commercial aircraft and presents a detailed statistical examination of ice-supersaturated air masses (RHice >100%). The analysis is confined to a region of high measurement density between latitudes 40°N- 60°N and longitudes 105°W and 30°E, for the years 1995-2010. Several conclusions are drawn regarding the probabilities of encountering regions of ice supersaturation (ISSR) in three different longitude regimes, based on distance from the tropopause and season. There is also a minor attempt to attribute interannual variations in these probabilities to the North Atlantic Oscillation (NAO).

#### MAJOR COMMENTS

**Reviewer:** *Uncertainties are calculated and presented (typically 1 standard deviation) for most mean values derived in this paper. However, the uncertainties are often ignored when interpreting the mean values and making quantitative statements about them. One example, in Lines 396-397: "... the average ISSR occurrence probability is 29% in the troposphere and increases to 34% when approaching the tropopause layer." Given that the standard deviations of these mean values are each at least \_9%, the two averages are not statistically different, and the claimed "increase" is not significantly different from zero. A second example is Figure 11, where a horizontal line (indicating no seasonality) can easily be drawn within the uncertainties in each panel that show "annual cycles". Therefore, the statement (Line 506), "For all regions, ISSR occurrence probabilities are highest in the winter/spring and lowest in summer ..." is not supported by these seasonal averages with their statistical uncertainties. In view of this, why are most uncertainties in this paper calculated and presented as 1 standard deviation when the vast majority of scientific uncertainties are reported as 95% confidence intervals (i.e., approximately 2 standard deviations for large sample sizes)?*

**Authors:** The standard deviations reported from airborne observations include not only the variability of observations caused by the noise of the instrument, but also the natural variability of the atmosphere when sampling from a fast-moving platform, and the interannual variability when averaging over a period of 15 years. Hence, we added the following statement to Section 2.1: "In our study, we use statistical analyses in the following manner: when assessing results from laboratory studies and calibration experiment based on reproducible observations, we apply the 2- $\sigma$  criterion for the 95% confidence level; when interpreting results from atmospheric observations which are taken from fast-flying airborne platforms and cover 15 years of observations, including their interannual and lateral variabilities, we report the mean values and respective 1- $\sigma$  standard deviations and state statistical significance or insignificance, respectively."

In the light of this introductory statement, we rephrased the interpretation of ISSR occurrence frequency as: "With reference to the thermal (dynamical) tropopause, the mean ISSR occurrence probability is 31% (38%) in the upper troposphere below the tropopause layer. The observed increases of mean ISSR occurrence probabilities towards the tropopause layer are below statistical significance. For both tropopause definitions, the standard deviation of observed ISSR fractions is largest for the lowest UT layer of the analysed atmospheric region and decreases with increasing altitude."

For the same reason, we did not modify Figures 12 and 13 (formerly 11 and 12) but report the mean values including their standard deviations.

**Reviewer:** *The "occurrence probability" statistics are simple to understand, based on the numbers of RH measurements reflecting subsaturation, saturation, or supersaturation during a flight segment, an entire flight, or a number of flights. But it is not clear how "occurrence probability standard deviation" statistics were calculated. Are these based on calculating an average of the occurrence probabilities for a number of flights, reflecting the variability of the occurrence probabilities for individual flights*



*around the average? This should be briefly explained, early in the paper, so the reader can immediately grasp the concept of the "occurrence probability standard deviation".*

**Authors:** We agree, that the procedure of calculating mean values and standard deviations from the data set is not sufficiently explained. Hence, we included a description in Section 2.1, reading: "Since each data set from one single flight provides only a one-dimensional snapshot of the state of the atmosphere along the flight track, and each aircraft cruises at a slightly different pressure level, the entire MOZAIC data are consolidated to season files of 3-months duration, allowing the analysis of vertical distributions of atmospheric state parameters on a robust statistical basis. For each season file, the statistical distribution (average and standard deviation, median and percentiles) of investigated properties (temperature, O<sub>3</sub> VMR, H<sub>2</sub>O VMR, RH<sub>ice</sub>, ISSR fraction) is calculated with respect to the above defined UT, TP and LMS vertical layers. From these seasonal averages or percentiles, respective 15-year mean values and standard deviations are determined."

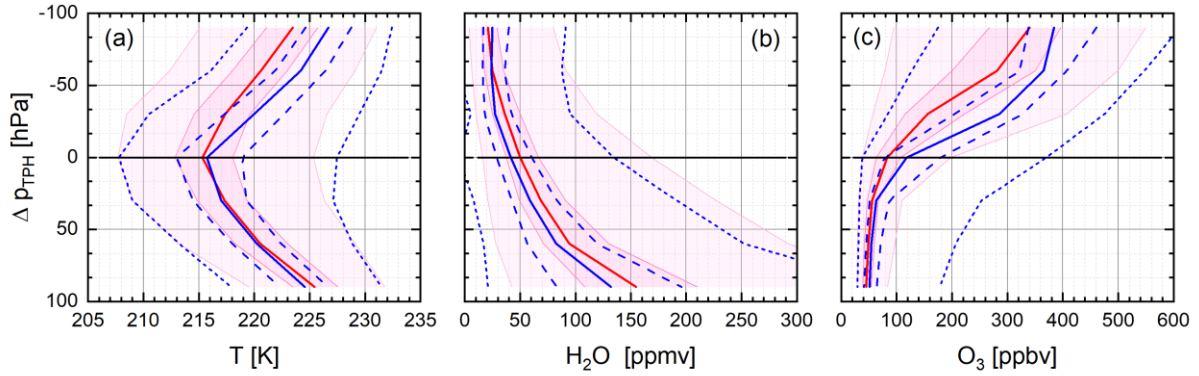
**Reviewer:** *Why is the requirement for supersaturation RH<sub>ice</sub> >100% when the measurement uncertainties are approximately 5% RH in the middle and upper troposphere? If some part of these uncertainties is a systematic error (a high bias of 3%, for example), wouldn't this lead to artificially high occurrence probabilities if measurements of a 98% RH air mass are 101% RH? How much do the occurrence probabilities decrease if you instead require RH<sub>ice</sub> >103%, or even RH<sub>ice</sub> >105% for supersaturation?*

**Authors:** As stated in Section 2.2, the MOZAIC capacitive humidity sensor MCH was extensively tested against research-grade instruments on the same platform, avoiding biases from sampling different air masses (Neis et al., 2015a; Neis et al., 2015b). The intercomparison study proved a zero offset with respect to RH<sub>ice</sub> data with a reported offset of  $-0.15 \pm 1.1.29\%$  RH<sub>L</sub> (Neis et al. 2015a). From these results we conclude that we don't see a systematic bias in the RH<sub>ice</sub> data reported from the MCH instrument. Hence, we assume a random scatter of the RH<sub>ice</sub> data around the zero value of RH<sub>ice</sub> = 100% which justifies the assumption of RH<sub>ice</sub> = 100% as threshold for ice-supersaturation.

**Reviewer:** *I'm not convinced that the comparison of supersaturation occurrence probabilities for atmospheric layers relative to the lapse rate ("thermal") tropopause vs the 2 PVU ("dynamical") tropopause shows much of a difference. If 95% confidence intervals of the mean values in Table 3 are considered, none of the "thermal" and "dynamical" averages for any atmospheric layer are statistically different. A lot of text, Figures and Tables are devoted to this comparison, and what does it show? Very little, in my opinion. Instead (or in addition), I'd prefer to see some assessment of the accuracy of the ERA-Interim tropopause heights that are absolutely critical to this paper. Since ozone mixing ratios were also measured as part of MOZAIC, and ozone can be used to define a "chemical" tropopause, can you compare ozone-defined tropopauses to the ERA-Interim tropopauses to evaluate at least the consistency of the latter? For example, if ERA-Interim puts the tropopause 1 km above the aircraft and the ozone mixing ratio is 1 ppm that indicates a large (>1 km) error in the tropopause height. I'm not suggesting a full-scale comparison, but rather some comparisons that illustrate the possible errors in tropopause heights.*

**Authors:** As we argue in the explanation of the values listed in Table 3, the ISSR fractions relative to the thermal and the dynamical tropopauses do not differ significantly for the analysed layers, with the exception of the layer LMS1. For the LMS 1 layer the mean ISSR fraction wrt the dynamical tropopause is 3.5 times the mean value wrt to the thermal tropopause. For the other layers, the difference is 1.5 or less. We are confident that our analysis presented in Section 3.3 contributes new insights to science.

Concerning the request for a detailed assessment of ERA-Interim tropopause heights, we agree that this is a valuable study. However, related work has been recently published by Reutter et al. (2020) in close collaboration with the MOZAIC/IAGOS team. Besides, we analysed the vertical distributions of temperature, H<sub>2</sub>O VMR and O<sub>3</sub> VMR over the North Atlantic. The new Figure 9 presents the results:



**Figure 9.** Vertical distribution of temperature T (a), H<sub>2</sub>O VMR (b), and O<sub>3</sub> VMR (c) relative to the 2 PVU dynamical tropopause and to the thermal tropopause; vertical distributions relative to the thermal tropopause are presented as percentiles [1, 25, 50, 75, and 99] by blue lines and relative to the 2 PVU tropopause conditions by red-shaded areas.

These results confirm the good agreement between the ERA-Interim thermal tropopause height indicated as  $\Delta p_{\text{TPH}} = 0$  hPa (blue lines), the lowest temperatures detected by MOZAIC at  $\Delta p_{\text{TPH}} = 0$  hPa, and the chemical tropopause, indicated by O<sub>3</sub> VMR = 120 ppbv at  $\Delta p_{\text{TPH}} = 0$  hPa. We consider these results as sufficient proof for the consistency of the used data set. A respective sentence was added to the manuscript: “In addition, the results confirm the good agreement between the ERA-Interim thermal tropopause height indicated by  $\Delta p_{\text{TPH}} = 0$  hPa (blue lines), the lowest temperatures detected at  $\Delta p_{\text{TPH}} = 0$  hPa, and the chemical tropopause, indicated by O<sub>3</sub> VMR = 120 ppbv at  $\Delta p_{\text{TPH}} = 0$  hPa, and thus the consistency of the used data set.”

**Reviewer:** *Water vapor mixing ratios are discussed in some sections of the paper and are shown in some Figures, but nowhere in the paper is there a description of how these were determined. Were they measured directly with different instruments (as implied in Line 17 of the abstract) or were they calculated from the RH measurements, requiring concomitant measurements of pressure and temperature with their associated uncertainties?*

**Authors:** Indeed, it was not explicitly mentioned how the H<sub>2</sub>O VMR values have been obtained. We introduced on sentence in Section 2.1: “The H<sub>2</sub>O VMR was finally calculated from the simultaneously measured RH<sub>ice</sub> and temperature data and from the pressure recordings of the aircraft avionic system.”

In order to avoid confusion, we also rephrased the abstract which reads now: “Observation data originate from regular and continuous long-term measurements on board of instrumented passenger aircraft in the framework of the European research program MOZAIC (1994 – 2010) which is continued as European research infrastructure IAGOS (from 2011). Data used in our study result from collocated observations of O<sub>3</sub>, RH<sub>ice</sub> and temperature, and H<sub>2</sub>O VMR deduced from RH<sub>ice</sub> and temperature data.”

The paragraph on the uncertainty of MCH water vapour measurements was also modified and reads now: “Kunz et al. (2008) who performed a statistical analysis of water vapour measurements from the SPURT campaigns between 2001 and 2003 by a Lyman- $\alpha$  photo-fragment fluorescence hygrometer (Zöger et al., 1999; Meyer et al., 2015) and MOZAIC water vapour data from the same period determined a limit of detection (LOD) of 10 ppmv for the MOZAIC sensor. Applying the same 2- $\sigma$  criterion (95% confidence level), we obtain a MCH limit of detection (LOD) of RH<sub>ice,LOD</sub> = 10% which again transfers into a minimum detectable H<sub>2</sub>O VMR of approx. 10 ppmv at typical mid-latitude upper troposphere conditions (T = 218K, p = 250 hPa); see also Neis et al. (2015a) for a detailed discussion. As is discussed by Smit et al. (2014), the uncertainty of the temperature measurement of the MCH sensor is included in the determination of the MCH RH<sub>ice</sub> uncertainty so

that the precision of H<sub>2</sub>O VMR data deduced from MCH RH<sub>ice</sub> data can be determined directly from the uncertainty of RH<sub>ice</sub> measurements. Overall, the 5% RH uncertainty leads to a decreasing precision of H<sub>2</sub>O VMR deeper in the stratosphere and implies a limited use of the MOZAIC H<sub>2</sub>O sensor in the stratosphere dominated by low RH<sub>ice</sub> and thus an increasing large uncertainty (Kunz et al., 2008)."

**Reviewer:** *There are some awkward and confusing sentences in the paper that could benefit from re-writing. I will point out a few of these below, but I suggest the paper be proofread by a native English speaker to clean up and clarify some sentences.*

**Authors:** A language check was performed and indicated sentences have been clarified.

#### MINOR COMMENTS

**Reviewer:** *Lines 23-25: This statement implies there is an increasing trend in summertime water vapor mixing ratios in the lowermost stratosphere, but no similar trend in RH<sub>ice</sub>. I don't think this is what you mean to say, rather that mixing ratios in this region are highest during summer months without corresponding maxima in RH<sub>ice</sub>. If this is the case, doesn't it imply that temperatures in this region are also highest during summer months?*

**Authors:** Indeed we observe highest temperatures in this region for the summer months as well, see Figure 8 of the discussion paper. Adopting your suggestion, we rewrote the sentence:

"Annual cycles of the investigated properties document highest H<sub>2</sub>O VMR and temperatures above the thermal tropopause in the summer months, whereas RH<sub>ice</sub> above the thermal tropopause remains almost constant in the course of the year

**Reviewer:** *L51: The term "tropopause layer" is used throughout this paper, but where is it geophysically defined? On page 5 you limit the TPL to "tropopause pressure \_ 15 hPa", but that's a definition that is neither common or geophysically-based. It would enlighten the reader to know why you chose these limits for the TPL.*

**Authors:** As explained in the general response to all reviewers we rewrote the introduction and refer to the extratropical transition layer, see line 55ff. Here, the references to the literature about the tropopause region and its structure are given. Concerning the pressure width of 30 hPa for the atmospheric layers around the tropopause, we refer to the commonly used approach when analysing MOZAIC data (Thouret et al., 2006).

**Reviewer:** *L64: Why would an "increase in pressure" change the RH of an air mass? RH is the partial pressure of water vapor divided by the saturation pressure over ice at a given temperature. Neither of these is affected in any way by an "increase in pressure". Please either explain this statement more clearly or remove it.*

**Authors:** As explained in details in Spichtinger and Leschner (2016), relative humidity can be expressed as  $RH_{ice} = p_q / (\epsilon p_{ice}(T))$ , using ideal gas assumptions; thus, ambient air pressure  $p$  and relative humidity  $RH_{ice}$  are linked by a linear relationship. However, the formation of ice-supersaturated regions is mostly driven by vertical uplift (i.e. decrease in temperature AND pressure), thus this possible effect is not probable and negligible.

We have rephrased the sentence as follows:

"Air masses supersaturated with respect to ice ( $RH_{ice} > 100\%$ ), so called ice-supersaturated regions (ISSR), have mostly faced a decrease in temperature or increase in water vapour mixing ratio, i.e. specific humidity during their past lifetime (Spichtinger and Leschner, 2016). As a result, these air parcels are both colder and of higher relative humidity than the embedded sub-saturated atmosphere (Gierens et al., 1999; Spichtinger et al., 2003b) which did not experience similar changes in their atmospheric state parameters."

**Reviewer:** L109-111: *Why are radiosonde network measurements of RH "considered insufficient for detecting trends and variability in UTLS water vapor"? I believe the GRUAN radiosonde data product for RH will be sufficient in this regard, and that GRUAN represents another existing global-scale network of in situ observations of atmospheric composition in the Ex-UTLS. A good reference for GRUAN is:*

*Bodeker, G.E., Bojinski, S., Cimini, D., Dirksen, R., Haeffelin, M., Hannigan, J., Hurst, D., Leblanc, T., Madonna, F., Maturilli, M., Mikalsen, A., Philipona, R., Reale, T., Seidel, D., Tan, D., Thorne, P., Vömel, H., and Wang, J.: Reference Upper Air Observations for Climate: From Concept to Reality, B. Am. Meteorol. Soc., 97, 123–135, <https://doi.org/10.1175/bams-d-14-00072.1>, 2016.*

**Authors:** Thank you for the recommendation. We have rephrased the paragraph to clarify the argument and added a specific sentence on GRUAN. The paragraph reads now:

"Concerning in-situ observations of water vapour, the international radiosonde network of weather balloons is in operation for many decades but the observations are considered insufficient for detecting trends and variability in UTLS water vapour; see Müller et al. (2016) and references therein. The GCOS Reference Upper-Air Network (GRUAN) targets the provision of climate-quality measurements of tropospheric and lower stratospheric variables (Seidel et al., 2009). GRUAN has established rigorous data quality assessment measures to provide reference-quality in situ and ground-based remote sensing observations of upper-air essential climate variables and serves as another source of high-quality water vapour data, however, for a limited number of certified surface stations yet (Bodeker et al., 2016)."

**Reviewer:** L180: *Please insert "attached" between "inlet" and "to"*

**Authors:** Done.

**Reviewer:** L184-189: *How about the uncertainty of RH measurements in the lower stratosphere? LOD is one measure, but since you determine supersaturation occurrence probabilities for several layers above the tropopause this must be somewhat known.*

**Authors:** We have expanded the discussion and added uncertainty ranges for different altitudes. The paragraph reads now:

"The MCH reports RH data with an average uncertainty of 4% RH (span 1% RH to 6% RH) in the middle troposphere at 4 to 8 km altitude during ascent and descent, and 5% RH (span 2% RH to 8% RH) at the tropopause and lowermost stratosphere at 10 to 12 km cruising altitude (Smit et al., 2014)."

**Reviewer:** L211: *Please change "sequences" to "segments". "Sequences" is also awkward in L105.*

**Authors:** Done.

**Reviewer:** Figure 3: *I think you intend "w/t" to mean "without" in both panels. Please change to "w/o".*

**Authors:** Done

**Reviewer:** L251-253: *Presumably Figure 4 shows the RH<sub>ice</sub> with the IFC applied, so please make this clear.*

**Authors:** The figure caption reads now: "Distribution of RH<sub>ice</sub> for the entire MOZAIC period from 1995 to 2010 with IFC applied as a function of ambient temperature with the colour indicating the probability of occurrence; the lines represent water saturation (solid line; Sonntag, 1994) and the threshold RH<sub>ice</sub> for homogeneous ice nucleation (dotted line; Koop et al., 2000; Kärcher and Lohmann, 2002)."

**Reviewer:** L286-287: *"highest possible quality achievable by this kind of routine observations" sounds great, but what does it actually mean? This sounds like an advertisement instead of a scientific claim and I suggest toning it down or removing it.*

**Authors:** We want to express that all possible corrections and consideration of potential sources of uncertainties have been made. We rephrased it to: “In summary, this data set is now considered of highest possible quality achievable by the type of sensor applied and for the type of routine observations performed.”

**Reviewer:** L290-297, Figure 6: *Up to this point, everything has focused on RH measurements and the tropopause-relative pressure bins you have defined. Here, the discussion suddenly turns to water vapor mixing ratios and tropopause-relative altitude bins. As above, where do the VMR data come from? And why does Figure 6 use altitude instead of pressure (or log pressure) as the vertical coordinate?*

**Authors:** The calculation of monthly mean profiles and the use of km as vertical spacing was done exclusively for producing Figs. 6 and 7 which can be compared directly to respective figures from Zahn et al. (2014). The main purpose is to illustrate the distinctly different vertical distributions of these two properties to the reader. The detailed information on the vertical distribution of related properties T, H<sub>2</sub>O VMW and RH<sub>ice</sub> is shown in Fig. 8.

The source of H<sub>2</sub>O VMR measurements is introduced in: “The H<sub>2</sub>O VMR was finally calculated from the simultaneously measured RH<sub>ice</sub> and temperature data and from the pressure recordings of the aircraft avionic system.”

**Reviewer:** L327: *“are bounded to the Great Lakes area and further North”. Given the size of Figure 1, it is difficult (without magnification) to find the Great Lakes. A better description would be “are within the northern half of the continental USA and southern half of Canada”.*

**Authors:** Good suggestion, done.

**Reviewer:** Figure 8, Tables 1 and 2: *It is not clear what the tropopause-relative pressure boundaries are for the different layers. Are the average values plotted (Figure) and presented (Tables) at  $\Delta p = -30$  hPa for the layer bounded by TPpress-15hPa and TPpress- 45hPa? This should be clearly stated.*

**Authors:** We clarified the vertical spacing of the layers in section 2.1:

“In order to reach both a sufficiently large data set for robust statistical analyses and good vertical resolution, the Ex-UTLS is subdivided into seven layers of 30 hPa thickness each, with three layers located below the thermal tropopause height and three layers above. Thouret et al. (2006) used a similar definition, but referenced to the dynamical tropopause at 2 PVU, i.e. they defined the tropopause as a mixing zone 30 hPa thick across the 2 PVU potential vorticity surface.

The seven layers of 30 hPa thickness each are centred at  $p_{\text{therm.TPH}} = 0$  hPa for the tropopause layer (TPL) itself and then at  $p_{\text{therm.TPH}} \pm 30$  hPa,  $p_{\text{therm.TPH}} \pm 60$  hPa, and finally at  $p_{\text{therm.TPH}} \pm 90$  hPa. From this vertical spacing, the separation of air masses is achieved by applying the following criteria (formulated for the thermal tropopause only):

LMS :  $p < p_{\text{therm.TPH}} - 15\text{hPa}$ ; which is limited by the maximum cruise altitude with  $p \approx 190$  hPa;

TPL :  $p = p_{\text{therm.TPH}} \pm 15\text{hPa}$ ;

UT :  $p > p_{\text{therm.TPH}} + 15\text{hPa}$ ; limited to lower altitudes by  $p < 350\text{hPa}$ .

**Reviewer:** *Given the standard deviations (Table 2), are the average values for different seasons or longitudinal regions (Table 1) statistically different at the 95% level of confidence?*

**Authors:** Following an argument of another reviewer, we replaced the standard deviations in Table 2 by the normalized standard deviations to make the values better comparable. Nevertheless, from a rigorous statistical point of view, the mean values for the different seasons are not significant at the 95% level of confidence. However, they reflect the interannual variability of the atmosphere since the seasonal means are calculated from 15 annual values. This interannual variability should not be neglected here.

**Reviewer:** L373: *Why is the annual cycle of UTH increasingly damped as you get closer to the tropopause?*

**Authors:** The sentence was removed.

**Reviewer:** L377: *As noted above, please explain how an increase in pressure can cause supersaturation.*

**Authors:** This statement has been removed.

**Reviewer:** L429-433: *This long sentence is confusing and requires re-wording for clarification.*

**Authors:** The section was divided into several sentences. It reads now:

“Above both tropopause layers, H<sub>2</sub>O VMR further decreases in case of non-ISSR conditions. For ISSR conditions, however, H<sub>2</sub>O VMR remains constant with height throughout the layer just above the tropopause. Doubling of H<sub>2</sub>O VMR for tropopause ISSR conditions compared to non-ISSR conditions is close to the results reported from MLS observations (Spichtinger et al., 2003b). In contrast, Gierens et al. (1999) found an increase of only 50% for H<sub>2</sub>O VMR inside ISSR compared to non-ISSR. In turn, this value compares well with our observations in the uppermost troposphere.”

**Reviewer:** Figure 9: *What does the black horizontal line represent at +70 hPa in the H<sub>2</sub>O panels?*

**Authors:** This was an error in the plotting routine. In all panels the line  $\Delta p = 0$  is highlighted as guideline to the eye for the tropopause layer.

**Reviewer:** L454-455: *The 33.5 ppb O<sub>3</sub> value from ERA-Interim is representative of what altitude and region?*

**Authors:** Cirisan et al. (2013) use a value of 33.5 ppbv from ERA Interim air mass trajectory analyses as the tropospheric background ozone value in the upper troposphere in midlatitudes. The respective explanation was added.

**Reviewer:** L460: *Why the sudden switch from P99 to P95, without explanation?*

**Authors:** The values are taken from literature. We explain now as follows (line 570): “Note that P95 refers here to the 95 percentile value of the analysed data ensemble, as taken from Cohen et al. (2018).”

**Reviewer:** Figure 10: *Error bars for each marker would clearly show if the troposphericity values are statistically different (or not).*

**Authors:** The troposphericity values are determined from median and 99 percentile values of the entire ensemble. Hence, we cannot associate an uncertainty to the troposphericity parameters. Instead they describe a property of the entire ensemble.

**Reviewer:** L500: *Here in the text you claim that Figure 11 shows results for the "top UT layer", but the caption for Figure 11 says "calculations were conducted for the two UT layers positions closest to the tropopause."*

**Authors:** This was an error in the manuscript and has been corrected accordingly in the text while the figure caption is correct.

**Reviewer:** L506-507: *This statement is not supported by the average values when their uncertainties are considered.*

**Authors:** The statement was removed.

**Reviewer:** L513: *I assume the Lindenberg radiosonde RH data has been corrected using the GRUAN-recommended corrections? It is important to say this because the reader may assume that uncorrected RH data from radiosondes are good enough (they are not!). You might also reference the paper describing corrections to the Vaisala RS92 data:*

Dirksen, R. J., Sommer, M., Immler, F. J., Hurst, D. F., Kivi, R., and Vömel, H.: Reference quality upper-air measurements: GRUAN data processing for the Vaisala RS92 radiosonde, *Atmos. Meas. Tech.*, 7, 4463–4490, <https://doi.org/10.5194/amt-7-4463-2014>, 2014.

**Authors:** The Lindenberg data and the respective analysis were taken from the publication by Spichtinger et al. (2003). Data originate from corrected RS80A routine radiosonde observations which were calibrated against collocated RS90 ascents. Details of the applied data correction procedure are described in the publication.

**Reviewer:** *Figure 12: I don't see any information about the layer or layers for which the results are shown. Please state this in the caption.*

**Authors:** As in Figure 11, the analysis refers to the two UT layers positioned closest to the tropopause. A respective statement is added to the figure caption.

**Reviewer:** *L549: typo "15y ears"*

**Authors:** Done.

**Reviewer:** *L530: Why only 15 months of Lindenberg radiosonde data? There are more than 9 years of GRUAN-corrected Vaisala RS92 RH data from Lindenberg.*

**Authors:** As explained above, we used the data from literature, as published by Spichtinger et al. (2003). A detailed intercomparison between IAGOS and GRUAN RH<sub>ice</sub> observations is for sure highly valuable but beyond the scope of this manuscript.

**Reviewer:** *L551: "fits well" is an overstatement since the DJF and MAM averages for Lindenberg lie outside the MOZAIC mean  $\pm$  1 standard deviation envelope.*

**Authors:** The statement was revised to: "The 15-months cycle from the radio soundings is covered by the 15 years climatology of ISSR occurrence from MOZAIC, but contributes only a snapshot compared to the 15-years' time series."

**Reviewer:** *L549: I don't know what a "first exemplary analysis" is. Please explain.*

**Authors:** The term "exemplary" means that we conducted a first analysis from a shorter data set to demonstrate the possibilities of the combined N<sub>ice</sub> – RH<sub>ice</sub> data set. We have now removed the term "exemplary" from the manuscript for clarity.

**Reviewer:** *L553: Trend analyses are performed on supersaturation occurrence probabilities based on which tropopause definition?*

**Authors:** We stated earlier in the manuscript that all analysis except the results presented in section 3.3 were conducted with respect to the thermal tropopause. For clarity we added a respective sentence to the manuscript: "The bases of our analyses were the seasonally averaged observations in the uppermost tropospheric layer (UT) with respect to the thermal tropopause, and the respective average seasonal cycles depicted in Figure 12."

**Reviewer:** *L579: "thus" must be a typo because it makes no sense in this sentence. Also, please change "long-term average values" to "long-term seasonal average values."*

**Authors:** Indeed there is a word missing. It reads now: "The de-seasonalised time-series thus show positive ...". The term "seasonal" was inserted as suggested.

**Reviewer:** *L581-582: Three significant figures for trends and their uncertainties is not justified when the uncertainty values are nearly as large as the trends themselves. Why present the 1 standard deviation uncertainties when, presumably based on 2 standard deviation uncertainties, you claim in the next sentence that none of the trends are significant?*

**Authors:** To our understanding, one of the key messages of the manuscript is that we do NOT find trends in ISSR occurrence. In that respect we believe that it is justified showing the time series of ISSR fractions in Fig. 14 (formerly 13) and the de-seasonalised time series of ISSR fraction in Fig. 15 (formerly 14).

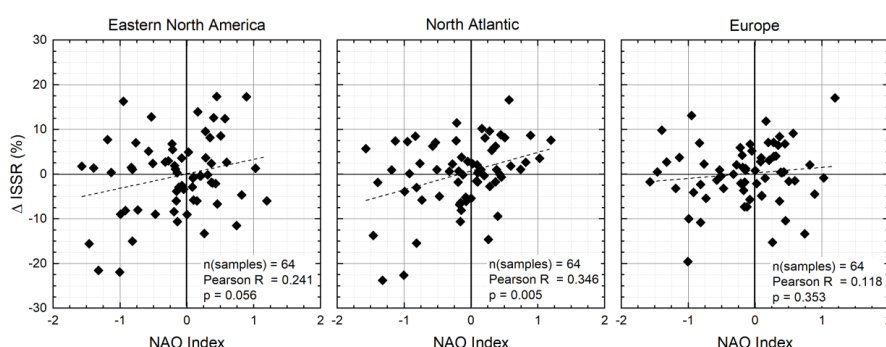
**Reviewer: L597:** *If the westerlies bring "warmer and more moist air to Europe", why would you expect a higher probability of supersaturation in the UT? More moisture increases the RH, but warmer air lowers the RH.*

**Authors:** The original wording was misleading. WE have rephrased the statement as: "This larger pressure difference causes stronger westerly winds and thereby more active storm tracks over the North Atlantic. Under such conditions we would expect a higher probability of ice-supersaturation in the uppermost troposphere due to more frequent warm conveyor belts that can induce the formation of ISSRs in the upper troposphere (Spichtinger et al., 2005)."

**Reviewer: L608:** *"we consider the correlation of signs statistically significant". This is a very qualitative conclusion that needs support from a quantitative explanation.*

**Authors:** The entire section was modified in response to a comment by Reviewer #1. We performed a cross-correlation analysis for all three regions and show the results in the modified Figure 16 (formerly 15). This cross-correlation analysis reported a statistically significant correlation between NAO index and deviation of ISSR occurrence from the long term average for the North Atlantic, but no significant correlations for the other two regions. The description of the figure and the conclusions drawn read now:

"For the regions Eastern North America and Europe the correlation between NAO index and  $\Delta$  ISSR fraction is not statistically significant. For the North Atlantic however, the results of the cross-correlation analysis indicate statistical significance at a level of 99%. The obtained correlation of signs is in line with the observation that the occurrence of ice-supersaturation is well correlated with the storm track activity (Spichtinger et al., 2003b; Gettelman et al., 2006; Lamquin et al., 2012)."



**Figure 16.** Correlation analysis with respect to the correlation of signs between NAO index and deviation of ISSR occurrence from the long-term average ( $\Delta$  ISSR) for the target regions; numbers indicate the results from the correlation analysis with respect to number of samples n, Pearson R and significance level p.

**Reviewer: L651:** *"which then generates more frequently ISSR" is awkward phrasing. Please rewrite.*

**Authors:** The sentence was rephrased: "This interpretation is in agreement with the understanding that a positive NAO index leads to an increased storm track activity which then may induce more frequent formation of ISSRs in the upper troposphere."



## Added References

Bodeker, G. E., Bojinski, S., Cimini, D., Dirksen, R. J., Haeffelin, M., Hannigan, J. W., Hurst, D. F., Leblanc, T., Madonna, F., Maturilli, M., Mikalsen, A. C., Philipona, R., Reale, T., Seidel, D. J., Tan, D. G. H., Thorne, P. W., Vomel, H., and Wang, J.: Reference upper air observations for climate: From concept to reality, *Bull. Amer. Meteorol. Soc.*, 97, 123-135, doi: 10.1175/bams-d-14-00072.1, 2016.

Diao, M., Zondlo, M. A., Heymsfield, A. J., Avallone, L. M., Paige, M. E., Beaton, S. P., Campos, T., and Rogers, D. C.: Cloud-scale ice-supersaturated regions spatially correlate with high water vapor heterogeneities, *Atmos. Chem. Phys.*, 14, 2639-2656, doi: 10.5194/acp-14-2639-2014, 2014.

Diao, M., Jensen, J. B., Pan, L. L., Homeyer, C. R., Honomichl, S., Bresch, J. F., and Bansemer, A.: Distributions of ice supersaturation and ice crystals from airborne observations in relation to upper tropospheric dynamical boundaries, *J. Geophys. Res.-Atmos.*, 120, 5101-5121, doi: 10.1002/2015jd023139, 2015.

Kunz, A., Spelten, N., Konopka, P., Mueller, R., Forbes, R. M., and Wernli, H.: Comparison of Fast In situ Stratospheric Hygrometer (FISH) measurements of water vapor in the upper troposphere and lower stratosphere (UTLS) with ECMWF (re)analysis data, *Atmos. Chem. Phys.*, 14, 10803-10822, doi: 10.5194/acp-14-10803-2014, 2014.

Neis, P.: Water Vapour in the UTLS - Climatologies and Transport, Forschungszentrum Jülich, Schriften des Forschungszentrums Jülich, Reihe Energie und Umwelt FZJ-2017-07862, 124 pp., 2017.

Pan, L. L., Bowman, K. P., Atlas, E. L., Wofsy, S. C., Zhang, F. Q., Bresch, J. F., Ridley, B. A., Pittman, J. V., Homeyer, C. R., Romashkin, P., and Cooper, W. A.: The Stratosphere-Troposphere Analyses of Regional Transport 2008 experiment, *Bull. Amer. Meteorol. Soc.*, 91, 327-342, doi: 10.1175/2009bams2865.1, 2010.

Reichler, T., Dameris, M., and Sausen, R.: Determining the tropopause height from gridded data, *Geophys. Res. Lett.*, 30, doi: 10.1029/2003gl018240, 2003.

Reutter, P., Neis, P., Rohs, S., and Sauvage, B.: Ice supersaturated regions: properties and validation of ERA-Interim reanalysis with IAGOS in situ water vapour measurements, *Atmos. Chem. Phys.*, 20, 787-804, doi: 10.5194/acp-20-787-2020, 2020.

# Ice-supersaturated air masses in the northern mid-latitudes from regular in-situ observations by passenger aircraft: vertical distribution, seasonality and tropospheric fingerprint

Andreas Petzold<sup>1</sup>, Patrick Neis<sup>1,3,§</sup>, Mihal Rütimann<sup>1</sup>, Susanne Rohs<sup>1</sup>, Florian Berkes<sup>1,§</sup>,  
5 Herman G.J. Smit<sup>1</sup>, Martina Krämer<sup>2,3</sup>, Nicole Spelten<sup>2</sup>, Peter Spichtinger<sup>3</sup>, Philippe Nedelec<sup>4</sup>,  
and Andreas Wahner<sup>1</sup>,

<sup>1</sup> Forschungszentrum Jülich GmbH, Institute of Energy and Climate Research 8 – Troposphere, Jülich, Germany

<sup>2</sup> Forschungszentrum Jülich GmbH, Institute of Energy and Climate Research 7 – Stratosphere, Jülich, Germany

<sup>3</sup> Johannes Gutenberg University, Institute for Atmospheric Physics, Mainz, Germany

10 <sup>4</sup> CNRS Laboratoire d'Aérodynamique, and Université Paul Sabatier Toulouse III, Toulouse, France

<sup>§</sup> now at CGI Deutschland B.V. & CO. KG, Frankfurt, Germany

<sup>§</sup> now at P3 solutions GmbH, Aachen, Germany

*Correspondence to:* Andreas Petzold (a.petzold@fz-juelich.de)

**Abstract.** The vertical distribution and seasonal variation of ~~upper tropospheric humidity (UTH)~~ water vapour  
15 volume mixing ratio ( $H_2O$  VMR), relative humidity with respect to ice ( $RH_{ice}$ ) and particularly of regions with  
ice-supersaturated air masses (ISSR) in the extratropical upper troposphere and lowermost stratosphere (~~Ex-~~  
~~UTLS)~~ is-are investigated at northern mid-latitudes over the regions Eastern North America, the North Atlantic  
and Europe for the period 1995 to 2010. Observation data originate from regular and continuous long-term  
measurements ~~of water vapour volume mixing ratio  $H_2O$  VMR, temperature and relative humidity with respect~~  
20 ~~to ice ( $RH_{ice}$ ) by on board of~~ instrumented passenger aircraft in the framework of the European research program  
MOZAIC (1994 – 2010) which is continued as European research infrastructure IAGOS (from 2011). Data used  
in our study result from collocated observations of  $O_3$  VMR,  $RH_{ice}$  and temperature, and  $H_2O$  VMR deduced  
from  $RH_{ice}$  and temperature data. The in-situ observations of ~~UTH- $H_2O$  VMR and  $RH_{ice}$~~  with a vertical  
resolution of 30 hPa (< 800 m at the extratropical tropopause level) and a horizontal resolution of 1 km resolve  
25 detailed features of the distribution of water vapour and ice-supersaturated air relative to the thermal tropopause,  
including their seasonal and regional variability and chemical signatures at various distances from the tropopause  
layer. Annual cycles of the ~~vertical distribution of UTH~~ investigated properties over the investigated regions  
demonstrate document annually increasing highest  $H_2O$  VMR and temperatures above the thermal tropopause in  
the summer months, whereas but without an associated increase in  $RH_{ice}$  above the thermal tropopause remains  
30 almost constant in the course of the year. Over all investigated regions, upper tropospheric air masses close to  
the tropopause level are nearly saturated with respect to ice and contain a significant fraction of ~~ice-~~  
~~supersaturated regions (ISSR)~~ with a distinct seasonal cycle of minimum values in summer (30% over the ocean,  
20 - 25% over land), and maximum values in late winter (35 - 40% over both land and ocean). Above the  
thermal tropopause, ISSR are occasionally observed with an occurrence probability of  $1.5 \pm 1.1\%$ , whereas above  
35 the dynamical tropopause at 2 PVU, the occurrence probability increases 4-fold to  $8.4 \pm 4.4\%$ . In both tropopause-

height (TPH) related coordinate systems, the ISSR occurrence probabilities drop to values below 1% for the next higher air mass layer with pressure levels  $p < p_{\text{TPH}} - 15$  hPa. For both tropopause definitions, the tropospheric nature or fingerprint, respectively, based on  $\text{O}_3$  VMR, indicate continuing ~~dominant~~-tropospheric influence on ISSR inside and above the respective tropopause layer. For the non-ISSR, however, the stratospheric nature is clearly visible above the thermal tropopause whereas above the dynamical tropopause the air masses show still a ~~relevant-substantial~~ tropospheric influence. For all three regions, seasonal deviations from the long-term annual cycle of ISSR occurrence show no significant trends over the observation period of 15 years, whereas a ~~statistically significant correlation between weak but significant dependencies of ISSR occurrence on~~ the North Atlantic Oscillation (NAO) index ~~and the deviation of ISSR occurrence from the long-term average are-is~~ observed ~~for the North Atlantic, but not for the continentally shaped regions Eastern North America and Europe~~.

## 1 Introduction

~~Upper tropospheric humidity (UTH)~~Relative humidity over ice and in particular ice-supersaturated air masses ( $\text{RH}_{\text{ice}} > 100\%$ ) are of ample importance for the occurrence and life cycle of high ice clouds, or cirrus clouds, respectively, which have a large but still not fully understood impact on Earth's climate, with its net radiation impact being unknown and even the sign being unclear (Chen et al., 2000; Boucher et al., 2013). In this context, long-term observations of ~~UTH-water vapour properties~~ are an indispensable prerequisite for the investigation of potential changes of ~~water-vapour~~ abundance in the global upper troposphere and lowermost stratosphere~~UTLS~~ (e.g., Müller et al., 2016), and the resulting effects on atmospheric radiation (e.g., Riese et al., 2012) ~~and-as well as~~ on cirrus cloud occurrence and life cycle (Gettelman et al., 2012; Krämer et al., 2016; Heymsfield et al., 2017).

The extratropical upper troposphere and lowermost stratosphere (Ex-UTLS) is characterised by thermal gradients and dynamical barriers which inhibit mixing, give rise to specific trace gas distributions and lead to a variety of definitions of the tropopause (Gettelman et al., 2011; Ivanova, 2013). The thermal tropopause according to WMO criteria (WMO, 1957) is defined as the level, where the lapse-rate decreases to  $2 \text{ K km}^{-1}$  or less and remains so small at least in the overlying layer of 2 km. This definition identifies the vertical change in the static stability and allows for the existence of multiple tropopause layers. The dynamical tropopause is based on the potential vorticity (PV) and includes both changes in static stability and vorticity (i.e., horizontal and vertical wind shear), also viewed as the dynamic stability. The PV values in the stratosphere exceed its values in the troposphere by an order of magnitude. The threshold value of 2 PVU ( $1 \text{ PVU} = 10^{-6} \text{ m}^2 \text{ K s}^{-1} \text{ kg}^{-1}$ ) for separating tropospheric and stratospheric air masses is commonly used in studies on stratosphere-troposphere transport. The chemical tropopause is based on the chemical change at the tropopause, identified from tracer-tracer correlations (Zahn and Brenninkmeijer, 2003), with a threshold value of  $\text{O}_3$  VMR  $> 120$  ppbv being used to distinguish stratospheric from tropospheric air (Thouret et al., 2006). The coexistence of different definitions of the tropopause and the observation that characteristics of air masses around the tropopause depend on the applied definition motivated the concept of the extratropical transition layer (ExTL) which describes the extratropical layer around the tropopause; see Gettelman et al. (2011) and references therein.

The vertical distribution of ~~water-vapour-trace species around-in~~ the Ex-UTLS is controlled by the strong static stability gradients and dynamic barriers to transport in this atmospheric layer. In the case of water vapour, the  $\text{H}_2\text{O}$  VMR is also determined by the coldest temperature the air parcel has experienced on its way to the

75 tropopause (Lagrangian dry/cold point), which decouples the abundance of water vapour from local cross-  
tropopause mixing to some extent (Hoor et al., 2010; Zahn et al., 2014). The distribution is described tropopause  
is characterized by a steep decrease of the H<sub>2</sub>O volume-mixing-ratio (VMR) up to the thermal-tropopause layer.  
Across the tropopause layer, H<sub>2</sub>O VMR decreases further but less steep until it reaches its near-constant  
stratospheric value at about 2 km altitude above ~~the tropopause layer~~. The thermal tropopause forms thus an  
80 efficient barrier for the large-scale vertical transport of H<sub>2</sub>O into the stratosphere, whereas troposphere-  
stratosphere transport occurs for specific local-scale dynamic situations such as, e.g., tropopause folds (Hoor et  
al., 2004; Hoor et al., 2010; Gettelman et al., 2011).

These features are reported from extensive research campaigns like SPURT (Hoor et al., 2004) which was  
designed on a climatological approach and compared to climatological data from the research programme  
85 MOZAIC (Marenco et al., 1998), and from long-term sampling by the CARIBIC passenger aircraft which carries  
an instrumented airfreight container (Dyroff et al., 2014; Zahn et al., 2014), or by instrumented balloons (Kunz  
et al., 2013). ~~The complex structure of the Ex-UTLS, and the extratropical transition layer are described in depth~~  
~~in the review article by.~~

Of particular interest with respect to ~~UTH and~~ ice cloud formation and life cycle is the thermodynamic state  
90 parameter RH<sub>ice</sub> which controls the properties of ice clouds by setting the thermodynamic conditions for cirrus  
cloud formation, existence and dissolution (Pruppacher and Klett, 1997). Air masses supersaturated with respect  
to ice (RH<sub>ice</sub> > 100%), so called ice-supersaturated regions (ISSR), have mostly faced a decrease in temperature;  
~~increase in pressure~~, or increase in water vapour mixing ratio, i.e. specific humidity during their past lifetime  
(Spichtinger and Leschner, 2016). As a result, these air parcels are both colder and ~~more humid~~ of higher relative  
95 humidity than the ~~embedding-embedded~~ sub-saturated atmosphere (Gierens et al., 1999; Spichtinger et al.,  
2003b) which did not experience similar changes in their atmospheric state parameters.

In the northern mid-latitudes, ISSR occurrence coincides strongly with the storm tracks over the North Atlantic  
(Spichtinger et al., 2003b; Gettelman et al., 2006; Lamquin et al., 2012). Frequently occurring synoptic weather  
features such as fronts or warm conveyor belts lead to synoptic-scale upward motion and thus facilitate the  
100 formation of ISSR (Spichtinger et al., 2005). However, ice-supersaturation, occurs as well in regions of high  
pressure and anticyclonic flow (Gierens and Brinkop, 2012). Detailed studies of the ISSR life cycle by means of  
Lagrangian trajectory analyses (Irvine et al., 2014) indicate that the lifetime of an air parcel in the state of  
supersaturation below the tropopause is generally short with the median duration being less than 6 hours for both  
winter and summer conditions. In an Eulerian view, however, these ISSR regions as composed of many  
105 supersaturated air parcels may persist on a much longer time scale (Spichtinger et al., 2005).

In contrast to the strong negative gradient in H<sub>2</sub>O VMR at altitudes below but close to the thermal tropopause,  
ISSR occur frequently in the humid and cold upper tropospheric air masses ~~close to the thermal tropopause~~.  
Detailed investigations of the distribution and structure of ice-supersaturation in the northern mid-latitude  
tropopause region over Lindenberg, Germany, from 15 months of balloon soundings showed that ice saturation  
110 occurs in most cases below the thermal tropopause, even in meteorological situations where the tropopause  
pressure is relatively high (Spichtinger et al., 2003a). On the other hand, the occurrence of ISSR above the  
thermal tropopause is very rare with a fraction of approx. 6% of the observations over Lindenberg, reporting ice-  
supersaturation above the thermal tropopause. Direct evidence of the occurrence of ice-supersaturation above but  
close to the thermal tropopause report a fraction of 2% from an earlier analysis of MOZAIC data (Gierens et al.,  
115 1999).

ISSR constitute potential formation regions for ice clouds, persistent contrails and contrail-cirrus. In these cold and humid air masses, natural cirrus clouds may form by heterogeneous or homogeneous freezing processes (Koop et al., 2000; Hoose and Möhler, 2012; Heymsfield et al., 2017), and long-lived contrails and contrail-cirrus are generated by cruising aircraft, causing the major non-CO<sub>2</sub> climate impact of civil aviation (Aaltonen et al., 2006; Stuber et al., 2006; Burkhardt et al., 2008; Lee et al., 2010; Burkhardt and Kärcher, 2011; Kärcher, 2018; Bock and Burkhardt, 2019).

The occurrence of ISSR and its close link to the occurrence of cirrus clouds is reported from a joint analysis of SAGE II data on subvisible cirrus and MOZAIC ice-supersaturation by Gierens et al. (2000) which provides an almost 1:1 relationship between subvisible cirrus occurrence and ice-supersaturation, but without discrimination between tropospheric and stratospheric air masses. From other platforms, there are only very few reports of cirrus clouds above the tropopause layer, either from satellite retrievals (Spang et al., 2015) or from research aircraft flights (Müller et al., 2015).

Despite the high climate-related relevance of the vertical distribution of water vapour VMR and related RH<sub>ice</sub> in the vicinity of the extratropical tropopause layer, there exist only very few approaches for the continuous global-scale monitoring of water vapour abundance and UTH-RH<sub>ice</sub> with sufficient precision and vertical resolution; see Müller et al. (2016) for an overview. Among space-borne techniques, the High-Resolution Infrared Radiation Sounder (HIRS) instruments are most important since they cover more than 3 decades of observations (Gierens et al., 2014), whereas the Microwave Limb Sounder (MLS) and the Atmospheric InfraRed Sounder (AIRS) were particularly used for the space-borne global mapping of ISSR (Spichtinger et al., 2003b; Lamquin et al., 2012) and cirrus cloud coverage (Stubenrauch et al., 2010). However, the vertical resolution provided by space-borne instruments in the Ex-UTLS is very limited and does not allow detailed studies on the vertical distribution of UTH-RH<sub>ice</sub> in this region.

Concerning in-situ observations of water vapour, the international ~~radiosonde~~-network of weather balloons is in operation for many decades but the observations are considered insufficient for detecting trends and variability in UTLS water vapour; see Müller et al. (2016) and references therein. The GCOS Reference Upper-Air Network (GRUAN) targets the provision of climate-quality measurements of tropospheric and lower stratospheric variables (Seidel et al., 2009). GRUAN has established rigorous data quality assessment measures to provide reference-quality in situ and ground-based remote sensing observations of upper-air essential climate variables and serves as another source of high-quality water vapour data, however, for a limited number of certified surface stations yet (Bodeker et al., 2016).

The only other existing global-scale in-situ observation infrastructure for atmospheric composition in the Ex-UTLS uses instrumented passenger aircraft for routine measurements of trace gases like H<sub>2</sub>O, O<sub>3</sub>, CO, greenhouse gases and nitrogen oxides, aerosols and clouds at cruise altitude. IAGOS (In-service Aircraft for a Global Observing System; see Petzold et al. (2015), Nédélec et al. (2015), and [www.iagos.org](http://www.iagos.org) for details) and its predecessor research programs MOZAIC (Marengo et al., 1998) and CARIBIC (Brenninkmeijer et al., 1999; Brenninkmeijer et al., 2007) conduct regular measurements of UTH since 1994. The transformation of both former research projects MOAZIC and CARIBIC into the current IAGOS Research Infrastructure took place in 2011. These regular flights on a global scale are unique in its quantity, continuity, and quality of measurements of Ex-UTLS air masses and have provided detailed insights into the distribution of RH<sub>ice</sub> (Gierens et al., 1999; Spichtinger et al., 2002), the distribution and properties of ISSR (Gierens and Spichtinger, 2000; Spichtinger and

Leschner, 2016), their link to cirrus clouds (Gierens et al., 2000; Petzold et al., 2017), and the processes controlling the water vapour distribution (Zahn et al., 2014).

In the present study, we analysed the distribution properties of  $RH_{ice}$  and of ISSR in the Ex-UTLS for a latitudinal band reaching from Eastern North America across the North Atlantic to Europe. We used the full

MOZAIC period from 1995 to 2010 which permits the robust seasonal analysis for the identified target regions.

Our studies focus on the structure of the vertical distribution of  $RH_{ice}$ , its variability and seasonality, and potential trends. The horizontal resolution of our data set is 1 km, set by the instrument time resolution of 4 s and the cruising speed of approx. 250 m s<sup>-1</sup>.

The vertical resolution ~~of our data~~ is set to 30 hPa, which corresponds to a vertical distance of approx. ~~800–750~~ m at ~~40 km~~ cruise altitude (Thouret et al., 2006) ~~for U.S. Standard~~

~~Atmosphere conditions~~ and assures sufficient statistical robustness of the conducted analyses. This vertical resolution ~~, and~~ is of similar order as the typical resolution of UTLS ~~models~~ data with a vertical grid spacing of about 50 hPa in the vicinity of the tropopause (Reichler et al., 2003). ~~General circulation~~ Chemistry-climate

models ~~of the middle atmosphere~~ like L90MA and L47MA use a vertical grid spacing of ~~40–2015–25~~ hPa near the extratropical tropopause (Jöckel et al., 2016) which is reflected in the selected vertical resolution of

MOZAIC data layers.

## 2 MOZAIC RH data set

### 2.1 Data coverage and vertical distribution

The MOZAIC RH data set used for this analysis spans over the period from 1995 to 2010 and is constrained to cruise altitude conditions, i.e., pressure below 350 hPa (above approx. 8 km altitude), and to ambient

temperatures below 233 K to exclude potential sensor contamination by supercooled liquid water droplets. The areal boundaries of the analysed data set are 40 °N to 60 °N and cover the regions Eastern North America (105 °W to 65 °W), North Atlantic (65 °W to 5 °W) and Europe (5 °W to 30 °E). Figure 1 illustrates the global

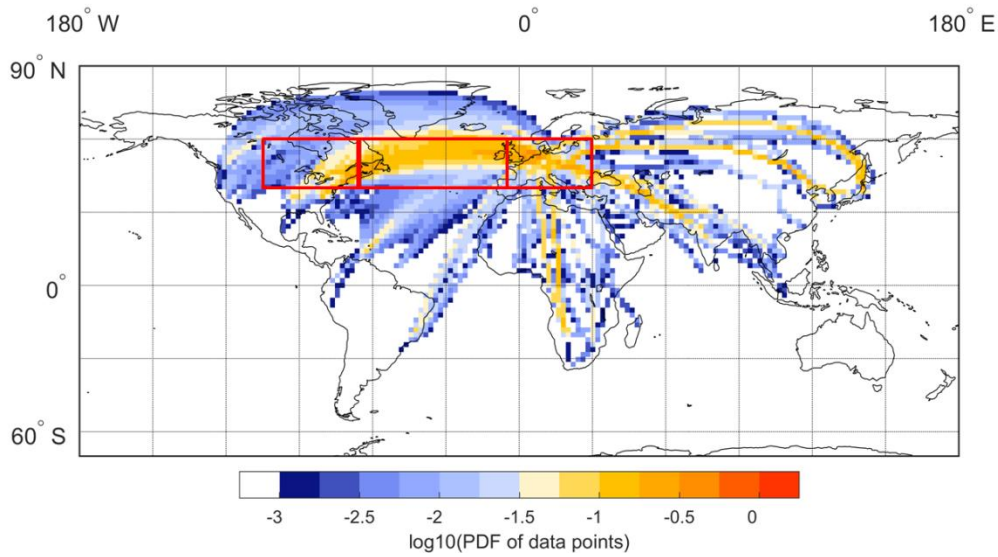
coverage of water vapour observations by MOZAIC for the years 1995 to 2010. Inserted boxes mark the regions Eastern North America, North Atlantic and Europe. The annual data coverage for ~~all each~~ analysed regional boxes

varies between 30 ~~hours~~ and 65 flight hours of flight-MOZAIC aircraft per season (3 months) which corresponds to 27,000 to 60,000 data points of 4 s duration each, per season per year. All investigated regions are characterized by continuous data coverage over the investigated period with no data gaps. Data are available to open access through the IAGOS data portal at [www.iagos.org](http://www.iagos.org).

Since MOZAIC data are collected at constant-pressure cruise levels of passenger aircraft which may cross from the upper troposphere (UT) through the tropopause layer (TPL) into the lowermost stratosphere (LMS) and back,

the data vertical coordinates are reported relative to the tropopause pressure level. ~~The extratropical tropopause layer height can be defined following different criteria (Gettelman et al., 2011; Ivanova, 2013). The thermal tropopause according to WMO criteria (WMO, 1957) is defined as the level, where the lapse rate decreases to~~

~~2 K km<sup>-1</sup> or less and remains so small at least in the overlying layer of 2 km. This definition, however, allows for the existence of multiple tropopause layers. The dynamical definition of the tropopause is based on the potential vorticity with 1 PVU = 10<sup>-6</sup> m<sup>2</sup> K s<sup>-1</sup> kg<sup>-1</sup>. The values of the potential vorticity in the stratosphere exceed its values in the troposphere by an order of magnitude. The threshold value of 2 PVU for separating tropospheric and~~



**Figure 1.** Global coverage of water vapour observations by MOZAIC for the period 1995 to 2010, shown as decadal logarithm of the probability distribution function (PDF) of the data points (fraction of measurements in a certain grid box); red boxes indicate the target areas for our analyses.

The pressure levels of the thermal tropopause ( $p_{\text{therm.TPH}}$ ) and the dynamical 2 PVU tropopause ( $p_{\text{dyn.TPH}}$ ) we are derived from ERA-Interim ~~data~~ (Dee et al., 2011) which uses 60 model layers with the top of the atmosphere located at 0.1 hPa. For our analysis, the 6-hourly outputs from ERA-I (0:75° x 0:75°) were interpolated onto a 1° x 1° horizontal grid and on 60 vertical levels of constant pressure and potential temperature (Kunz et al., 2014; Berkes et al., 2017). Additionally, the variables of the PV, and the pressure of the thermal tropopause ( $p_{\text{therm.TPH}}$ ) based on the WMO criteria were calculated (WMO, 1957; Reichler et al., 2003). The ERA-Interim data were then linearly interpolated with respect to longitude, latitude, pressure, and time onto each flight track with 4 s resolution, as described by Kunz et al. (2014). Interpolated tropopause pressure levels were finally used to determine the position of the aircraft relative to ~~this layer~~ the thermal tropopause or to the 2 PVU iso-surface, respectively, and thus to distinguish whether the aircraft sampled air masses of UT, TPL or LMS origin with respect to the chosen tropopause definition.

In order to reach both a sufficiently large data set for robust statistical analyses and good vertical resolution, the Ex-UTLS is subdivided into seven layers of 30 hPa thickness each, with three layers located below the thermal tropopause height and three layers above. Thouret et al. (2006) used a similar definition, but referenced to the dynamical tropopause at 2 PVU, i.e. they defined the tropopause as a mixing zone 30 hPa thick across the 2 PVU potential vorticity surface.

The seven layers of 30 hPa thickness each are centred at  $p_{\text{therm.TPH}} = 0$  hPa for the tropopause layer (TPL) itself and then at  $p_{\text{therm.TPH}} \pm 30$  hPa,  $p_{\text{therm.TPH}} \pm 60$  hPa, and finally at  $p_{\text{therm.TPH}} \pm 90$  hPa. From this vertical spacing, ~~t~~he separation of air masses is achieved by applying the following criteria (formulated for the thermal tropopause only):

LMS :  $p < p_{\text{therm.TPH}} - 15\text{hPa}$ ; which is limited by the maximum cruise altitude with  $p \approx 190$  hPa;

TPL :  $p = p_{\text{therm.TPH}} \pm 15\text{hPa}$ ;

UT :  $p > p_{\text{therm.TPH}} + 15\text{hPa}$ ; limited to lower altitudes by  $p < 350\text{hPa}$ .



The bulk of our analyses refer to the classic thermal tropopause according to WMO criteria (WMO, 1957), with the exception of the occurrence of ISSR above the tropopause, where we present the analyses for both tropopause definitions and compare the results [to learn more about the processes influencing the formation of ISSR](#); see Section 3.3.

~~In order to reach both a sufficiently large data set for robust statistical analyses and good vertical resolution, the Ex-UTLS is subdivided into 7 layers of 30 hPa thickness each, with three layers below the TPL and 3 layers above. Thouret et al. (2006) used a similar definition, but referenced to the dynamical tropopause at 2 PVU.~~

~~A more detailed description of the methodology for determining the aircraft position relative to the thermal tropopause and for the vertical resolution of the data set is given by Berkes et al. (2017).~~ Since each data set from one single flight provides only a one-dimensional snapshot of the state of the atmosphere along the flight track, and each aircraft cruises at a slightly different pressure level, [the entire MOZAIC data are consolidated to season files of 3-months season-filesduration](#), allowing the analysis of vertical distributions of atmospheric state parameters on a robust statistical basis. [For each season file, the statistical distribution \(average and standard deviation, median and percentiles\) of investigated properties \(temperature, O<sub>3</sub> VMR, H<sub>2</sub>O VMR, RH<sub>ice</sub>, ISSR fraction\) is calculated with respect to the above defined UT, TP and LMS vertical layers. From these seasonal averages or percentiles, respective 15-year mean values and standard deviations are determined.](#)

[In our study, we use statistical analyses in the following manner: when assessing results from laboratory studies and calibration experiments based on reproducible observations, we apply the 2- \$\sigma\$  criterion for the 95% confidence level; when interpreting results from atmospheric observations which are taken from fast-flying airborne platforms and cover 15 years of observations, including their interannual and lateral variabilities, we report the mean values and respective 1- \$\sigma\$  standard deviations and state statistical significance or insignificance, respectively.](#)

## 2.2 RH and O<sub>3</sub> instrumentation

The relative humidity measurements of MOZAIC and today IAGOS use a thin-film capacitive sensor of type Humicap (Vaisala) which is mounted inside an aeronautic Rosemount inlet [attached](#) to the aircraft skin. The MOZAIC Capacitive Hygrometers (MCH) are calibrated in the laboratory against a Lyman  $\alpha$  resonance fluorescence hygrometer (Kley and Stone, 1978) with respect to RH over liquid water (Helten et al., 1998; Smit et al., 2014). The conversion to RH<sub>ice</sub> uses the equations by Sonntag (1994). The MCH reports RH data with an [average uncertainty of 4% RH \(span 1% RH to 6% RH\) in the middle troposphere at 4 to 8 km altitude during ascent and descent, and 5% RH \(span 2% RH to 8% RH\) at the tropopause and lowermost stratosphere at 10 to 12 km cruising altitude](#) (Smit et al., 2014). [The H<sub>2</sub>O VMR was finally calculated from the simultaneously measured RH<sub>ice</sub> and temperature data and from the pressure recordings of the aircraft avionics system.](#)

The deployed sensor has been carefully compared to high-precision water vapour instruments in dedicated research aircraft studies (Helten et al., 1999; Neis et al., 2015a; Neis et al., 2015b) which demonstrate a remarkably good agreement between the MCH and reference instruments with  $R^2 = 0.92$  and a slope of  $m = 1.02$  from linear regression analyses. The authors report an MCH uncertainty of 5% RH which is in close agreement with the uncertainty determined from error propagation analysis (Smit et al., 2014).

~~Applying the 2- $\sigma$  criterion (95% confidence level), the MCH limit of detection (LOD) is RH<sub>ice,LOD</sub> = 12% which transfers into a minimum detectable H<sub>2</sub>O VMR of approx. 10 ppmv at typical mid-latitude upper troposphere conditions (T = 218K, p = 250 hPa).~~ Kunz et al. (2008) who performed a statistical analysis of water vapour



measurements from the SPURT campaigns between 2001 and 2003 by a Lyman- $\alpha$  photo-fragment fluorescence

265 hygrometer (Zöger et al., 1999; Meyer et al., 2015) and MOZAIC water vapour data from the same period  
~~applied a similar~~ determined a limit of detection (-LOD) value of 10 ppmv for the MOZAIC sensor. Applying the  
same 2- $\sigma$  criterion (95% confidence level), we obtain a MCH limit of detection (LOD) is of  $RH_{ice, LOD} = 102\%$   
which again transfers into a minimum detectable  $H_2O$  VMR of approx. 10 ppmv at typical mid-latitude upper  
troposphere conditions ( $T = 218K$ ,  $p = 250$  hPa); see also Neis et al. (2015a) for a detailed discussion. As is  
270 discussed by Smit et al. (2014), the uncertainty of the temperature measurement of the MCH sensor is included  
in the determination of the MCH  $RH_{ice}$  uncertainty so that the precision of  $H_2O$  VMR data deduced from MCH  
 $RH_{ice}$  data can be determined directly from the uncertainty of  $RH_{ice}$  measurements. Overall, the 5% RH  
uncertainty leads to a decreasing precision of  $H_2O$  VMR deeper in the stratosphere and implies a limited use of  
the MOZAIC  $H_2O$  sensor in the stratosphere dominated by low  $RH_{ice}$  and thus an increasing large uncertainty  
275 (Kunz et al., 2008).

Since the launch of MOZAIC, the programme provides also  $O_3$  VMR data, in addition to  $H_2O$  and  $RH_{ice}$   
observations. Aboard MOZAIC and now IAGOS aircraft, ozone is measured by means of a UV absorption  
instrument which is characterised by an instrument noise of  $\pm 2$  ppbv and an integration time of 4 s (Nédélec et  
al., 2015). We used the collocated measurement of  $O_3$  and  $H_2O$  /  $RH_{ice}$  for the characterisation of ice-  
280 supersaturated air masses with respect to a potential stratospheric influence.

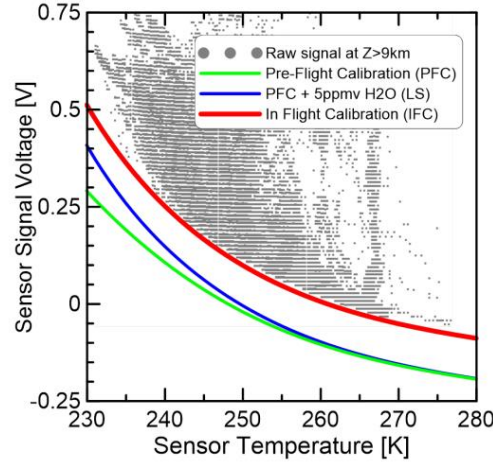
### 2.3 RH data processing

The processing of the MCH data had been subject to a calibration error from year 2000 on. This error in the data  
analysis caused a bias of data towards higher  $RH_{ice}$  values and shifted the peak value of the  $RH_{ice}$  probability  
distribution function (PDF) for in-cloud observations to approx. 130%  $RH_{ice}$  which is far above the physically  
285 expected value of 100%  $RH_{ice}$ . Earlier MCH data for the period 1995 to 1999, however, are not affected. The  
publications by Lamquin et al. (2012) ( Fig. 5 of that publication) and Penner et al. (2018) (Fig. 6 of that  
publication) illustrate the shift of the erroneous MOZAIC data towards higher  $RH_{ice}$  values very clearly.

The calibration error was corrected in a recent reanalysis and the PDFs of  $RH_{ice}$  are now consistent for the full  
MOZAIC period and physically reasonable with the PDF showing a second maximum at 100%  $RH_{ice}$ , as  
290 expected for in-cloud ~~sequences-segments~~ (Smit et al., 2014).

Besides the calibration error, another limitation of the MOZAIC RH data set stemming from MCH sensor drifts,  
required correction. In its standard operation mode, MCH sensors were replaced every 3 to 6 months. During  
their deployment periods, the sensors showed occasionally drifts of the sensor output signal caused by a shift of  
the sensor offset voltage, which results in erroneously high  $RH_{ice}$  values. To overcome this measurement artefact,  
295 the so-called in-flight calibration method (IFC) was developed by Smit et al. (2008), which references the offset  
voltage of the sensor to signals from flight ~~sequences-segments~~ in dry stratospheric air masses where the  
expected  $RH_{ice}$  signal is below the MCH LOD and thus the true MCH signal is considered zero RH.

The methodology is illustrated in Figure 2: The MCH sensors leave the calibration facility with a baseline for  
dry conditions (green curve); the theoretical signal expected from the stratospheric  $H_2O$  background of 5 ppmv is  
300 then added and this new baseline (blue curve) is the reference line for the offset determination. In the operational  
mode of the IFC method, the lower bound values of the MCH signal during an operational period of typically 15  
consecutive flights are determined as the observations below the 1 Percentile value (P01) of the data collected

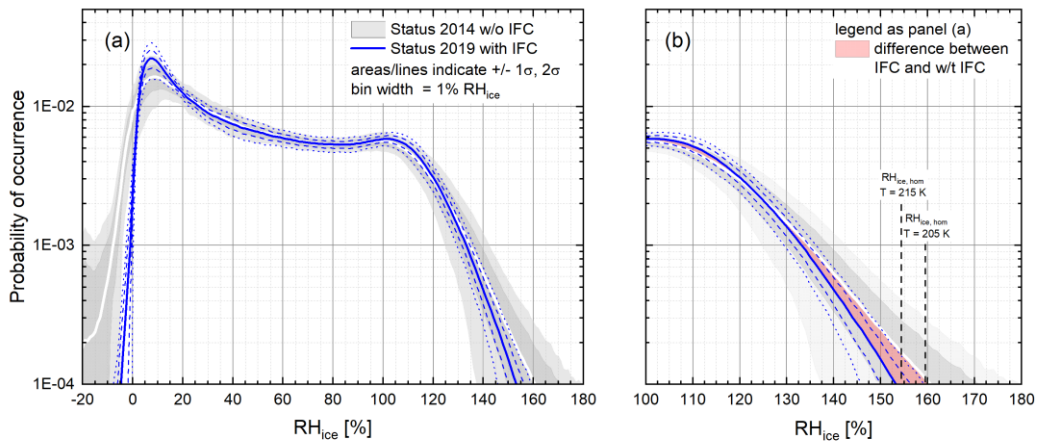


**Figure 2.** Raw signal of the MOZAIC humidity sensor aboard one MOZAIC aircraft as a function of the sensor temperature inside the aeronautic housing obtained at cruise altitude ( $z = 9 - 12$  km). Green line: zero signal from pre-flight calibration (PFC); blue line: superposition of zero signal from PFC and contribution by 5 ppmv water vapour; red line: zero signal from In-Flight Calibration (IFC).

during the respective flight sequence. In case of a sensor offset drift during MCH operation, the lower envelope from the P01 values is similar to the baseline for dry conditions at calibration plus the 5 ppmv stratospheric  $H_2O$  background value, but shifted by a voltage offset. The difference between the lower envelope and the baseline from calibration determines the sensor offset voltage which is then subtracted from the raw signal. Details of the methodology are described in detail by Smit et al. (2008).

## 2.4 RH data validation

The IFC method was applied to the full reanalysis data set from 1995 to 2010. Figure 3a illustrates the effect of the IFC method for the averaged  $RH_{ice}$  PDF from for the entire MOZAIC period data set, irrespective of the geographical regions where the data were collected. The presented average PDF and variability is calculated from annual PDFs.

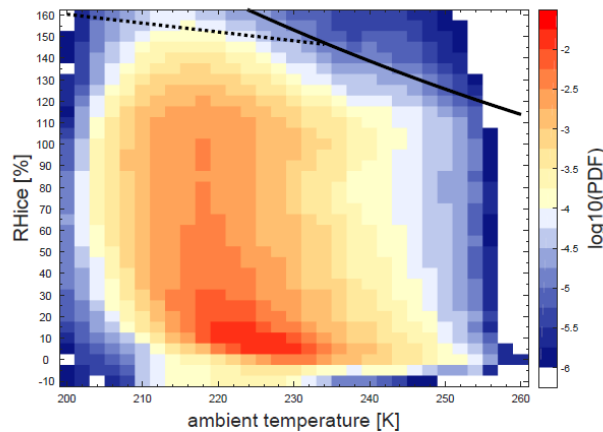


**Figure 3.** Averaged probability density functions of  $RH_{ice}$  for the entire MOZAIC period from 1995 to 2010 (a), and the zoom into the region of ice-supersaturation (b); data stem from the reanalysis (Smit et al., 2014) without (white line, grey areas), and with (blue lines) the in-flight calibration method applied to the data; red-shaded area

indicates the difference between IFC applied (with IFC) and not applied (w/o IFC), and vertical lines indicate the threshold  $RH_{ice}$  values for homogeneous nucleation of ice at  $T = 205$  K and  $T = 215$  K (Koop et al., 2000).

Solid lines refer to the MOZAIC average PDF without the IFC method (white) and with the IFC method applied (blue). Grey areas (without IFC) and dashed and dotted blue lines (with IFC applied) represent the  $\pm 1\sigma$  and  $\pm 2\sigma$  ranges. Figure 3b shows a zoom into the PDF for the range with  $RH_{ice} > 100\%$ . In addition to Panel (a), the red area marks the difference between the averaged PDFs without and with IFC applied.

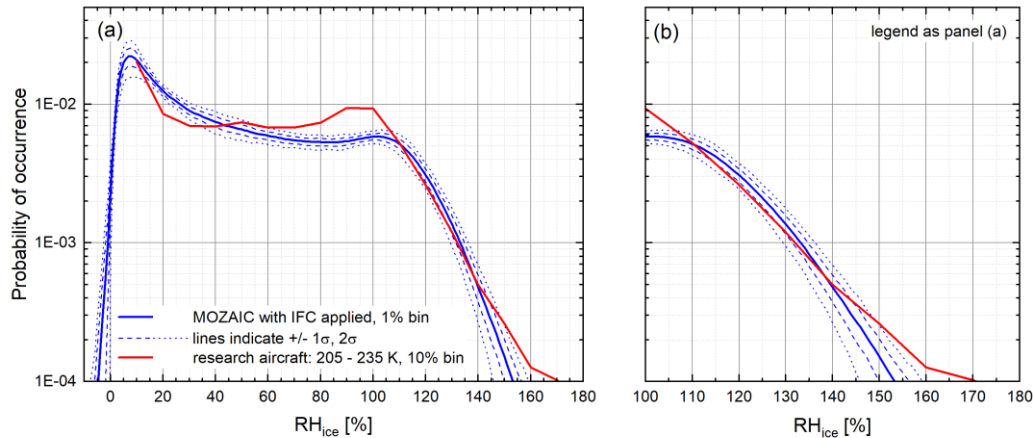
The overall features of the  $RH_{ice}$  PDF with an overall maximum value at dry stratospheric air mass values with  $RH_{ice}$  being close to the LOD of approx. 10%, and a second local maximum at  $RH_{ice} \approx 100\%$  for observations inside cirrus clouds remain unaffected, whereas the deviation between the average PDFs becomes relevant for  $RH_{ice}$  values above 130%. Here, the IFC leads to an average reduction of  $< 5\%$   $RH_{ice}$  for an occurrence probability of  $10^{-3}$  and approx. 7.5%  $RH_{ice}$  for an occurrence probability of  $10^{-4}$ . More relevant, the  $2\sigma$  - variability of the observed ice-supersaturations at  $10^{-4}$  occurrence probability reduces from max. 180%  $RH_{ice}$  (without IFC applied) to 155%  $RH_{ice}$  (with IFC applied). The latter value with the IFC applied fits into the range of the homogeneous freezing thresholds at typical extratropical tropopause conditions of  $RH_{ice,hom} = 158.25\%$  at 205 K to  $RH_{ice,hom} = 154.15\%$  at 215 K (Koop et al., 2000), as sampled by MOZAIC. Respective values without the IFC applied, however, exceed the homogeneous nucleation threshold significantly. Figure 4 illustrates the distribution of  $RH_{ice}$  observations from the entire MOZAIC data set shown in Figure 1 as a function of ambient temperature, colour-coded by the probability of occurrence, i.e. the fraction of data points for a specific combination of temperature and  $RH_{ice}$  with the respect to the entire ensemble. Obviously,  $RH_{ice}$  observations remain inside the physical boundaries let by the water saturation line and the line for homogeneous ice nucleation.



**Figure 4.** Distribution of  $RH_{ice}$  for the entire MOZAIC period from 1995 to 2010 with IFC applied as a function of ambient temperature with the colour indicating the probability of occurrence; the lines represent water saturation (solid line; Sonntag, 1994) and the threshold  $RH_{ice}$  for homogeneous ice nucleation (dotted line; Koop et al., 2000; Kärcher and Lohmann, 2002).

Besides the validation of MOZAIC  $RH_{ice}$  distributions with respect to the homogeneous nucleation thresholds from (Koop et al., 2000), the data were compared to the distribution of  $RH_{ice}$  from observations on board of research aircraft by high-precision water vapour instruments such as Lyman- $\alpha$  photo-fragment fluorescence hygrometers (Zöger et al., 1999; Sitnikov et al., 2007), tunable diode laser absorption spectrometers (May and

Webster, 1993; Krämer et al., 2009; Buchholz et al., 2013), and frost point hygrometers; see Meyer et al. (2015) for details. In total, 250 research flights from 32 field campaigns conducted between 1999 and 2017 globally were analysed. To ensure comparability to the MOZAIC data set, the temperature range was restricted to 205 K to 235 K which corresponds to the MOZAIC observation range, with the upper temperature limit set by the homogeneous freezing threshold.



**Figure 5.** Averaged probability density functions of  $RH_{ice}$  for the entire MOZAIC period from 1995 to 2010; with the in-flight calibration method applied (blue lines) and respective  $RH_{ice}$  PDF from 250 research aircraft flights collected in the Juelich In-situ Airborne Database (Krämer et al., 2016).

The result of this validation-comparison is shown in Figure 5. The MOZAIC  $RH_{ice}$  PDF are-is plotted similar to Figure 3, whereas the  $RH_{ice}$  PDF from the research aircraft campaigns is shown as red line, calculated for  $RH_{ice}$  bin widths of 10%. Both probability distribution functions show excellent agreement within the uncertainty ranges, particularly for the regime of ice-supersaturation (panel b). The differences for  $RH_{ice}$  near 100% are caused by the preferred sampling of ice clouds during the field campaigns (higher probability of ice clouds at  $RH_{ice} \approx 100\%$ ) and by frequent sampling of contrails at subsaturated conditions ( $RH_{ice} < 100\%$ ).

$RH_{ice}$  observations from the CARIBIC passenger aircraft exhibit similar features as the observations shown here from MOZAIC and from research aircraft, with maximum probability of occurrence at  $RH_{ice} = 100\%$  and maximum  $RH_{ice}$  values of approx. 150% (Dyroff et al., 2014). In that respect, all observation platforms provide consistent information on the distribution of ice-supersaturation in the extratropical tropopause.

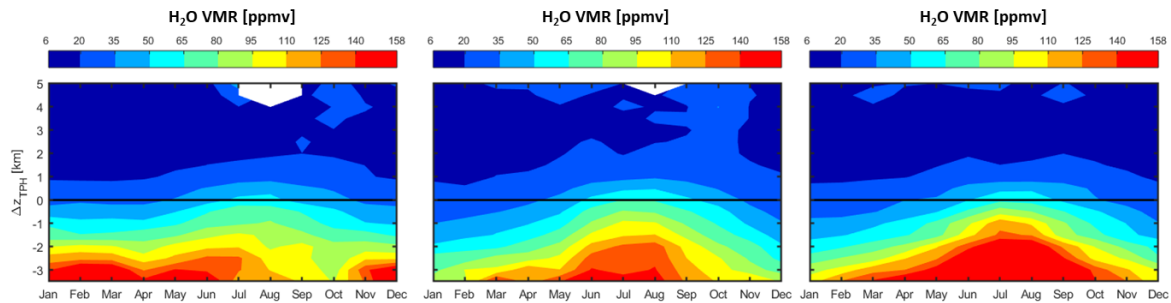
With the IFC method applied to the full MOZAIC  $RH_{ice}$  data, this data set is successfully validated against  $RH_{ice}$  observations by high-precision instruments and against physically justified bounding values. In summary, this data set is now considered of highest possible quality achievable by the type of sensor applied and for the this kind-of-type-of routine observations performed.

### 3 Results

#### 3.1 Annual cycles of water vapour and $RH_{ice}$ distributions at the tropopause

The annual cycles of the vertical distributions of water vapour volume mixing ratio ( $H_2O$  VMR) and  $RH_{ice}$  were analysed for the three target regions Eastern North America (ENA), North Atlantic (NAtl) and Europe (EU),

based on 15-year averages of monthly mean profiles relative to the thermal tropopause. For all investigated regions, the annual cycles of H<sub>2</sub>O VMR vertical distributions are shown in Figure 6. For the lowest layer of the lowermost stratosphere, bounded from below by the thermal tropopause layer, the patterns are similar for the three regions, characterised by low H<sub>2</sub>O VMR values in winter and spring months and a maximum H<sub>2</sub>O VMR during summer. For all regions, the influence of upper tropospheric air masses reaches approx. 1.0 - 2.0 km above the tropopause, with strongest influence in summer.



**Figure 6.** 15-year averaged annual cycles of H<sub>2</sub>O VMR vertical distributions of H<sub>2</sub>O VMR for latitudes 40 °N to 60 °N and for the regions (from left to right) Eastern North America (105 °W to 65 °W), North Atlantic (65 °W to 5 °W) and Europe (5 °W to 30 °E).

Below the tropopause layer, however, we find different behaviour for the studied regions. It appears that over the North Atlantic and over Europe which is strongly influenced by the North Atlantic synoptic weather systems due to the prevailing westerly winds, the annual cycles of H<sub>2</sub>O VMR in the uppermost troposphere and tropopause layers are coupled, while for the Eastern North American region the upper free troposphere layers seem to exhibit higher specific humidity ~~be more humid~~ in winter than respective air masses over the ocean. At the tropopause level however, the differences vanish and the annual cycles converge.

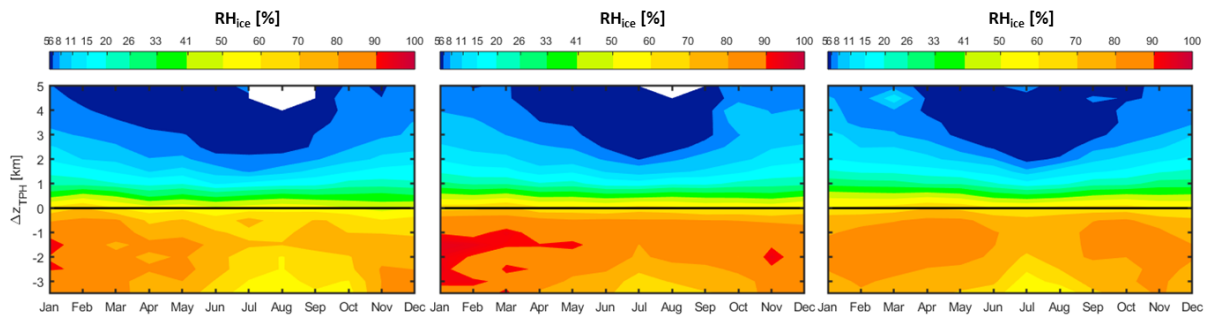
A similar behaviour of the annual cycle of H<sub>2</sub>O VMR was reported by Zahn et al. (2014) from zonal-averaged H<sub>2</sub>O VMR observations by the CARIBIC system. In contrast to MOZAIC, the CARIBIC H<sub>2</sub>O sensor provides good data also for the lower stratosphere where the MOZAIC RH sensor loses its sensitivity, but due to its limited regional coverage, the CARIBIC data set cannot provide regional-scale resolution. In that respect, these data sets complement each other with CARIBIC observations backing up the MOZAIC H<sub>2</sub>O VMR reported for the atmospheric layers just above the thermal tropopause and MOZAIC providing regional-scale resolution of seasonal patterns which is not possible otherwise.

Potential transport pathways of water vapour into the lowermost stratosphere are not in the scope of this study, and cannot be deduced from the analysis shown in Figure 6, but are discussed in depth elsewhere; see e.g., Gettelman et al. (2011), Zahn et al. (2014) and references given therein. In summary, the seasonal variation of H<sub>2</sub>O in the first 1-2 km above the tropopause is controlled by shallow, fast, two-way cross-tropopause mixing which is active around the year and is responsible for the extratropical tropopause mixing layer, or ExTL, respectively (Hoor et al., 2004), localized deep convection events which occur mainly in the summer period over continents (Anderson et al., 2012; Schwartz et al., 2013), and the hemisphere-scale effect of the Asian summer monsoon (Santee et al., 2017; Rolf et al., 2018). Strong cases of the deep convection events have been reported particularly for the Central United States with unusually wet conditions in the lowermost stratosphere being associated to these events (Anderson et al., 2017). Our long-term data do not point at a significantly higher humidity over the Eastern North America region in summer compared to the North Atlantic and to Europe.

However, it has to be noted that our observations are ~~bounded to the Great Lakes area and further North~~ within the northern half of the continental USA and the southern half of Canada (see Figure 1 for the areal coverage of MOZAIC observations), whereas the deep convection events with strong overshooting are reported for regions further ~~South~~ south over the Great Plains. This regional difference may explain the differing observations.

### 3.2 Annual cycles of $RH_{ice}$ and ISSR distributions at the tropopause

~~The core part of our~~ Our study is focusing on the vertical distribution, seasonality and regional variability of  $RH_{ice}$  and ice-supersaturated regions in particular which are linked to the water vapour content of the investigated atmospheric layers. Therefore, we discussed the observed water vapour distribution patterns in the preceding section. To shift the focus on  $RH_{ice}$ , Figure 7 represents a similar analysis as shown in Figure 6, but for relative humidity with respect to ice. In contrast to the differing annual cycles of water vapour distributions at the tropopause as discussed above, we find similar patterns for UTH and  $RH_{ice}$  over all target regions, with a tropopause layer characterised by mean  $RH_{ice}$  of 60% almost independent of the season, a very humid layer just below the tropopause with mean  $RH_{ice}$  reaching 80% and weak seasonality, and a stronger seasonality of UTH at approx. 1 km below the tropopause and further down into the upper free troposphere with dryer air during the summer season and very humid conditions particularly during winter and spring. Similar average values of  $RH_{ice}$  of 60–70% for the uppermost troposphere without significant seasonality are reported from CARIBIC observations (Dyroff et al., 2014; Zahn et al., 2014).



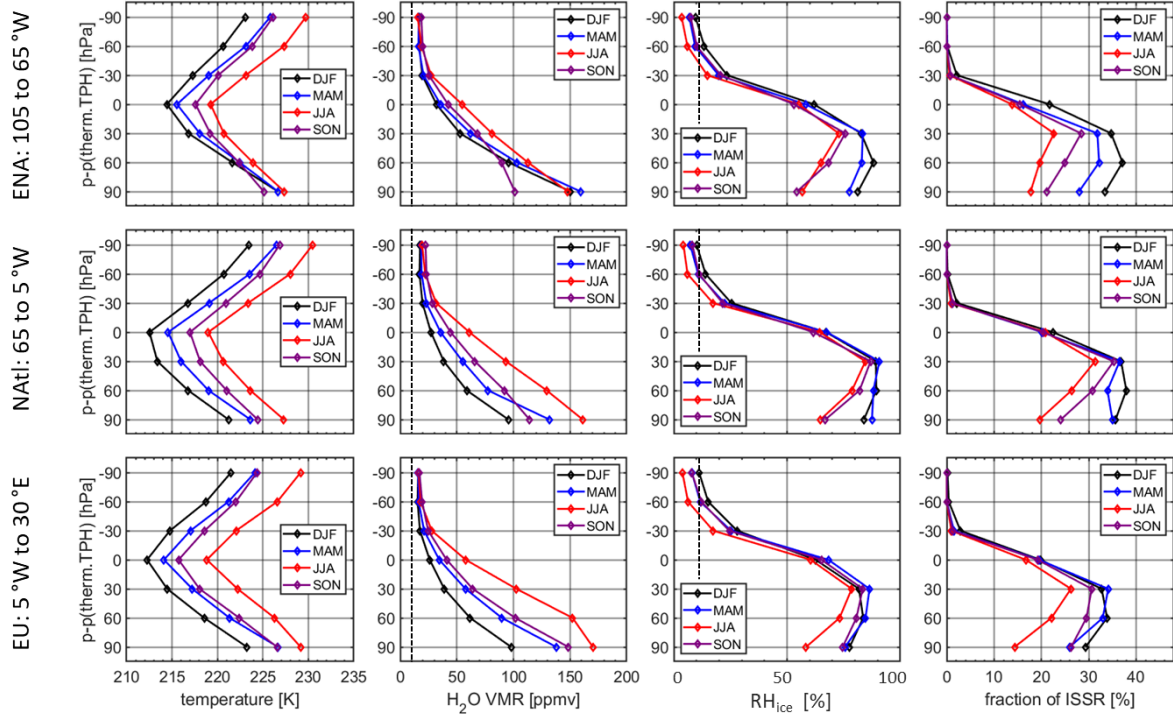
**Figure 7.** 15-year averaged annual cycles of  $RH_{ice}$  for latitudes 40 °N to 60 °N and for the regions (from left to right) Eastern North America (105 °W to 65 °W), North Atlantic (65 °W to 5 °W) and Europe (5 °W to 30 °E).

Grouping the data set shown in Figure 8 into seasonal clusters of layers of 30 hPa thickness around the tropopause allows the robust statistical analysis of the vertical distributions of temperature,  $H_2O$  VMR, average  $RH_{ice}$  and fraction of ice-supersaturated regions. The applied concept of the vertical spacing is described in Section 2.1. The seasonal variation of the vertical distributions of the selected properties is compiled in Figure 8. Table 1 and Table 2 present the average-mean fractions (Table 1) and associated standard deviations normalised to the respective mean values (Table 2) for ISSR occurrence, separated for regions and seasons, and in the last set of columns set-averaged over all regions. As is already indicated ~~by~~ in Figure 7, the variation of  $RH_{ice}$  with altitude and season is similar for the three target regions.

For all regions, the highest  $RH_{ice}$  values and also the highest fraction of ISSR occurrence is observed for the two upper tropospheric layers closest to the tropopause layer whereas for the third layer situated deepest inside the UT,  $RH_{ice}$  values and ISSR fractions are considerably lower. Only in the spring season (MAM) over the North



Atlantic, the lowest third layer reaches similar values for  $RH_{ice}$  values and ISSR fractions as the two layers above. Interestingly Focussing on the UT layers, the relative standard deviations of the ISSR fractions mean values are highest for the lowest layer investigated here, at least for winter and spring seasons for which the largest ISSR fractions is are found. The decline in variability with increasing altitude illustrates the damping of the annual cycle of UTH when getting closer to the tropopause layer.



**Figure 8.** Vertical distribution of mean temperature,  $H_2O$  mixing ratio,  $RH_{ice}$  and fraction of ice-supersaturated regions (ISSR) for seven pressure layers around the thermal tropopause; layer thickness is 30 hPa and layers are spaced equally relative to the tropopause pressure level; dotted lines indicate the MCH 2- $\sigma$  limit of detection of  $RH_{ice,LOD} = 12\%$  and the resulting minimum-detectable  $H_2O$  VMR of approx. 10 ppmv.

**Table 1.** ISSR frequency of occurrence-probability: seasonal average-mean values are reported in %; the vertical distance to the thermal tropopause is reported as  $\Delta p = p_{layer} - p_{therm.TPH}$ .

$\Delta p$ (hPa)	DJF			MAM			JJA			SON			AVG(ENA, Natl, EU)			
	ENA	Natl	EU	ENA	Natl	EU	ENA	Natl	EU	ENA	Natl	EU	DJF	MAM	JJA	SON
-30	2.1	2.2	2.9	0.7	1.2	1.5	0.6	0.9	1.0	0.8	1.2	1.3	2.4	1.1	0.8	1.1
0	21.7	22.4	19.8	16.2	20.1	19.5	13.8	20.8	16.7	15.5	20.4	19.2	21.3	18.6	17.1	18.4
30	34.7	36.8	32.8	31.8	36.5	34.1	22.7	31.3	26.3	28.4	35.3	30.6	34.8	34.2	26.8	31.4
60	37.1	37.9	33.9	32.2	34.0	33.0	19.6	26.4	22.1	25.0	30.8	29.5	36.3	33.0	22.7	28.5
90	33.5	35.7	29.3	28.0	35.0	26.0	17.7	19.6	14.3	21.1	24.1	26.3	32.8	29.6	17.2	23.8

**Table 2.** ISSR frequency of occurrence-probability: normalised standard deviations of seasonal average-mean values are reported in %; the vertical distance to the thermal tropopause is reported as  $\Delta p = p_{layer} - p_{therm.TPH}$ .

$\Delta p$	DJF	MAM	JJA	SON	AVG(ENA, Natl, EU)
------------	-----	-----	-----	-----	--------------------

(hPa)	ENA	NAtl	EU	ENA	NAtl	EU	ENA	NAtl	EU	ENA	NAtl	EU	DJF	MAM	JJA	SON
-30	102%	72%	84%	78%	47%	47%	99%	57%	50%	78%	33%	43%	86%	57%	68%	51%
0	26%	31%	31%	31%	21%	22%	31%	24%	19%	24%	16%	23%	29%	25%	25%	21%
30	15%	20%	15%	19%	18%	15%	31%	25%	23%	15%	10%	13%	17%	18%	26%	13%
60	19%	24%	20%	25%	17%	12%	31%	22%	22%	17%	13%	15%	21%	18%	25%	15%
90	29%	31%	25%	21%	33%	15%	19%	18%	31%	29%	15%	17%	29%	23%	23%	20%

### 3.3 Physico-chemical signature of ice-supersaturated regions in the vicinity of the tropopause

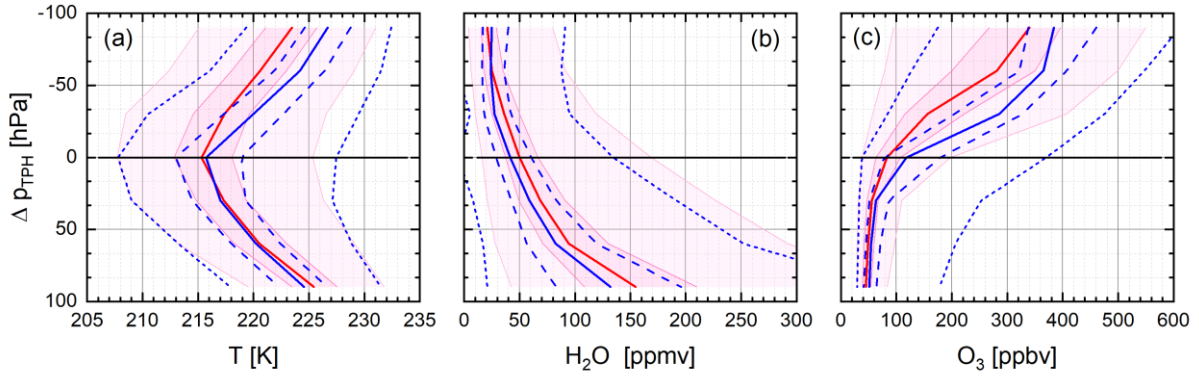
As discussed in detail by Spichtinger and Leschner (2016) ice-supersaturated air masses have mostly faced decrease in temperature, ~~increase in pressure~~, or increase in water vapour mixing ratio, i.e. specific humidity, during their past lifetime. Thus, these air parcels are known as both colder and more humid than the embedding sub-saturated air masses (Gierens et al., 1999; Spichtinger et al., 2003b). This conclusion is valid for both ISSR in the uppermost troposphere as well as for the rarer cases of ISSR above the tropopause.

In order to study the formation history of ISSR and involved processes, we analysed the occurrence frequency and physico-chemical signature of ISSR around the tropopause layer and referred our analyses to both the thermal and the dynamical tropopause. We want to recall the tropopause definitions given in Section 2.1. The thermal tropopause according to WMO criteria (WMO, 1957) is usually seen as an effective transport barrier hampering troposphere-stratosphere exchange, whereas the dynamical tropopause is commonly used for separating tropospheric and stratospheric air masses in studies on stratosphere–troposphere transport since it

represents the lower bound of the ~~tropopause mixing layer~~ ExTL. These complementary views on the tropopause have been developed from extensive CO - O<sub>3</sub> analyses, which showed that the 2 PVU surface approximately separates the troposphere from the stratosphere with the ExTL as a transition layer of about 2 km thickness above it and centred on the thermal tropopause (Hoor et al., 2004; Pan et al., 2010; Gettelman et al., 2011). These tracer studies in the extratropics showed that on average the dynamical tropopause is situated slightly below the thermal tropopause and the gradients of CO and O<sub>3</sub> are much sharper across the thermal tropopause compared to the dynamical tropopause (Hoor et al., 2004; Pan et al., 2010).

Similar features are observed for the gradients of temperature T, H<sub>2</sub>O VMR and O<sub>3</sub> VMR, shown in Figure 9 for the North Atlantic region. Similar to the tracer gradients, also the temperature gradient is sharper across the thermal tropopause compared to the dynamical tropopause. In addition, the results confirm the good agreement between the ERA-Interim thermal tropopause height indicated by  $\Delta p_{TPH} = 0$  hPa (blue lines), the lowest temperatures detected at  $\Delta p_{TPH} = 0$  hPa, and the chemical tropopause, indicated by O<sub>3</sub> VMR = 120 ppbv at  $\Delta p_{TPH} = 0$  hPa, and thus the consistency of the used data set. Furthermore, the analysis of the pressure difference between the thermal and dynamical tropopauses reveal an offset of approx. 25 hPa (15 - 35 hPa) which translates into an altitude difference of approx. 1 km (Neis, 2017).





**Figure 9.** Vertical distribution of temperature  $T$  (a),  $H_2O$  VMR (b), and  $O_3$  VMR (c) relative to the 2 PVU dynamical tropopause and to the thermal tropopause; vertical distributions relative to the thermal tropopause are presented as percentiles [1, 25, 50, 75, and 99] by blue lines and relative to the 2 PVU tropopause conditions by red-shaded areas.

Our analysis of ISSR occurrence in the vicinity of the exTL is confined to the North Atlantic region, for which we have the highest data density available with respect to vertical resolution. As described generally in Section 2.1, the entire data set of individual  $RH_{ice}$  observations over the North Atlantic region was divided into yearly subsets for seasons DJF, MAM, JJA, and SON. For each year, season and altitude layer relative to the thermal and dynamical tropopauses, the average frequency of occurrence of observations with  $RH_{ice} > 100\%$  was determined. For each season of the 15 years period we calculated the average occurrence probability for ISSRs per altitude layer relative to the thermal tropopause. The PDF probability of ISSR occurrence per altitude layer with respect to the entire period of 15 years was then calculated from this record of seasonally averaged ISSR frequencies of occurrence ensemble. For the sake of statistical significance, we skipped the distinction between seasons for this specific analysis and calculated instead median values and respective percentiles for the entire North Atlantic data set. However, seasonal information is contained in the statistical entity via the seasonally averaged ISSR fractions. The results are compiled in Table 3 for both tropopause definitions used here. Please note that the ISSR fractions compiled for the thermal tropopause correspond to the values listed in Table 2, but without distinction for seasons.

With reference to the thermal (dynamical) tropopause, the average mean ISSR occurrence probability is 29.31% (38%) in the upper troposphere and increases to 34% when approaching below the tropopause layer. With reference to the dynamical tropopause, the overall behaviour is similar with an The observed increases increasing average of mean ISSR occurrence probabilities  $y$  when reaching towards the tropopause layer are below statistical significance, but the absolute values are larger since the analysed layers reach deeper into the upper troposphere. For both tropopause definitions, the variability standard deviation of observed ISSR fractions is largest for the lowest UT layer of the analysed atmospheric region and decreases with increasing altitude.

**Table 3.** Mean and standard deviation of seasonal fraction of ice supersaturated regions (ISSR) for the seven vertical layers distributed around the thermal and dynamical tropopause.

Layer ID	$p - p_{TPH}$ [hPa]	ISSR fraction [%]
----------	---------------------	-------------------

		Dynamical TP	Thermal TP
LMS3	- 90	0.2±0.5	0.0±0.1
LMS2	- 60	0.7±1.1	0.1±0.3
LMS1	- 30	8.4±4.4	1.5±1.1
TPL	0	30.7±9.4	20.0±6.5
UT1	30	39.9±10.0	33.9±9.0
UT2	60	37.7±10.7	31.4±9.2
UT3	90	35.5±14.3	29.1±12.1

When crossing the thermal tropopause, the ISSR fraction drops sharply to values of 1.5% for the lowest layer above the thermal tropopause and to statistically insignificant fractions when reaching further up into the stratosphere. In case of the dynamical tropopause, we find a significantly higher ISSR fraction of 8.4% for the lowest stratosphere layer, and again insignificant fractions further above. This strong contrast in the ISSR occurrence probability for the lowest stratosphere layers with reference to the two tropopause definitions ~~is caused by the different physical natures of the thermal and dynamic tropopauses. coincides with the behaviour of other tracers in the ExTL; see Figure 9 for details.~~

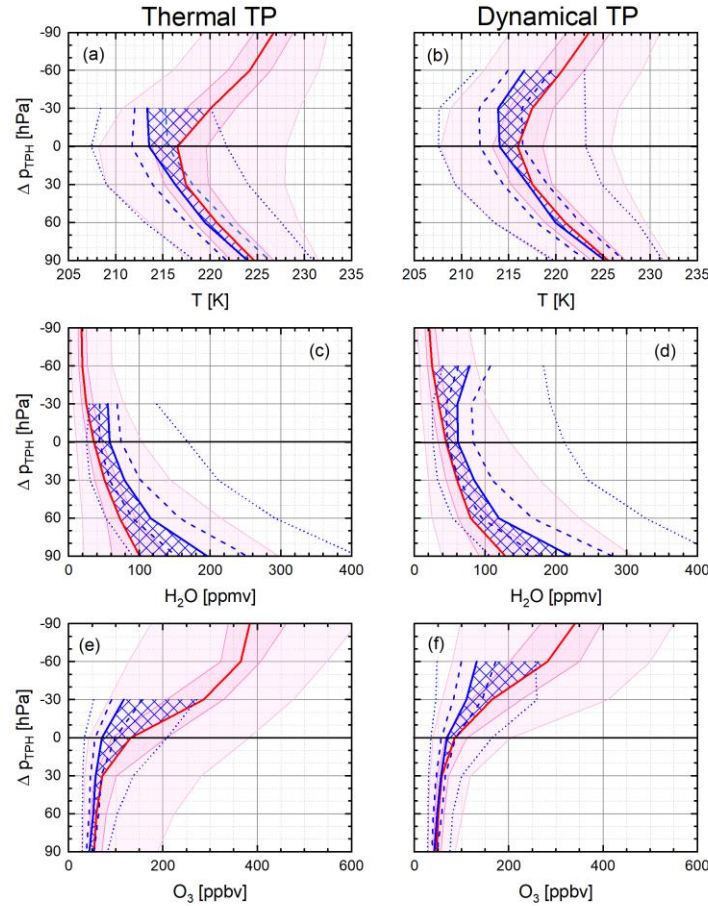
~~While the thermal tropopause forms a robust barrier for the vertical transport of water vapour, the dynamic tropopause serves as the lower bound for an atmospheric layer characterised by dynamically driven mixing processes. As a consequence, we expect different chemical signatures for the ISSR above the thermal and dynamical tropopauses.~~

In order to learn more about the history of ice-supersaturated air parcels we further analysed the ozone content of the ISSR compared to the sub-saturated air around, for air parcels below and above the thermal and dynamic tropopauses and combined the results with the distributions of temperature and H<sub>2</sub>O VMR. The thermodynamic and chemical properties of ISSR and the comparison between ISSR (blue lines) and ice-sub-saturated air masses (red-shaded areas and red lines) are presented in Figure 10 with reference to both tropopause definitions. In general, ISSR are colder than their subsaturated counterparts. The difference is low in the UT with 1 - 2 K which compares well to the value of 2 K at 215 hPa obtained from MLS satellite measurements (Spichtinger et al., 2003b), and increases to more than 6 K difference in the stratosphere above the thermal tropopause, and approx. 4 K above the dynamical tropopause. The temperature difference of 3 - 4 K between colder tropospheric ISSR and the surrounding subsaturated air masses reported by Gierens et al. (1999) is comparable to the temperature difference in the 30 hPa thick tropopause layer we find in our analysis.

Figure 10 also indicates a similar behaviour of the vertical distribution of H<sub>2</sub>O VMR for ice-supersaturated and ice-sub-saturated regions with exponentially decreasing absolute humidity up to the tropopause layer. Above both tropopause layers, H<sub>2</sub>O VMR ~~is~~ further decreases ~~for their case of~~ non-ISSR conditions. For ISSR conditions, however, H<sub>2</sub>O VMR, whereas the water vapour VMR remains constant with height ~~at the tropopause layer value of about 55 ppmv in the case of ISSR throughout the layer~~ just above the tropopause. ~~The increase~~ Doubling of H<sub>2</sub>O VMR ~~in for the~~ tropopause ISSR conditions compared to non-ISSR conditions by more than a factor of 2 is ~~comparable close~~ to the results ~~reported from~~ of MLS observations (Spichtinger et al., 2003b), ~~while In contrast,~~ Gierens et al. (1999) ~~reported found~~ an increase of only 50% for H<sub>2</sub>O VMR ~~water vapour VMR inside~~ ISSR compared to non-ISSR. In turn, this value ~~which~~ compares well ~~to with~~ our observations in the uppermost troposphere.

560 The vertical distribution of the ozone VMR behaves similar to the temperature for ice-supersaturated and ice-sub-saturated regions, with small differences in the ozone VMR of less than 15 ppbv in the troposphere. Already for the tropopause layer and even more pronounced for the first layer above the thermal tropopause, however, the difference increases to 60 ppmv ozone VMR and beyond.

565 Quantitative conclusions on air mass characteristics and history are drawn from the vertical distributions of thermodynamic and chemical properties shown in Figure 10. The underlying concept of troposphericity (Cirisan et al., 2013) quantifies the tropospheric nature or fingerprint, respectively, of an air mass on the basis of the observed  $O_3$  VMR. In the context of our study, we refer to troposphericity for consistency with literature.

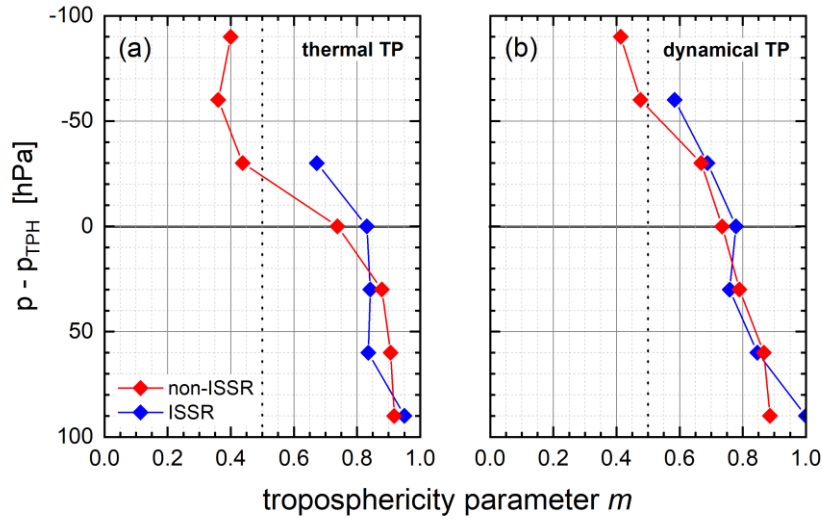


**Figure 10.** Vertical distribution of temperature,  $H_2O$  VMR, and ozone VMR for ISSR relative to the thermal tropopause height (panels a, c and e) and 2 PVU dynamical tropopause height (panels b, d, and f). ISSR conditions are presented as percentiles [1, 25, 50, 75, and 99] by blue lines and non-ISSR conditions by red-shaded areas; blue cross-hatched areas highlight the deviation of median values inside ISSR from those non-ISSR conditions.

575 | Using the ~~ozone volume mixing ratio~~ $O_3$  VMR as a stratospheric air mass tracer and adapting the ~~if~~ approach of Cirisan et al. (2013), we define the troposphericity parameter  $m$  for an ensemble of data characterised by median (med) and 99 percentile (P99) values as

$$m = \frac{[O_3]_{P99} - [O_3]_{med}}{[O_3]_{P99} - [O_3]_{tropo}}$$

and apply the median value of the lowest layer analysed here as background tropospheric value, so that  $[O_3]_{\text{tropo}}$   
 580 = 42 ppbv. Petetin et al. (2018) reported a median  $O_3$  VMR of 49 ppbv for the Central European mountain  
 station Sonnblick (3106 m above sea level) in the Austrian Alps and a value 50 ppbv for the high Alpine station  
 Jungfraujoch (3580 m above sea level), whereas Cirisan et al. (2013) use a value of 33.5 ppbv from ERA Interim  
 air mass trajectory analyses [as the tropospheric background ozone value in the upper troposphere in midlatitudes](#).  
 Applying this definition of the troposphericity parameter  $m$  to MOZAIC/IAGOS observations over Central  
 585 Europe (Petetin et al., 2018) at 4000 m altitude with  $[O_3]_{\text{med}} = 50$  ppbv and  $[O_3]_{\text{P99}} = 82$  ppbv yields  $m = 0.80$ ,  
 and for observations at 1500 m altitude with  $[O_3]_{\text{med}} = 42$  ppbv and  $[O_3]_{\text{P99}} = 83$  ppbv we find  $m = 1.00$ .  
 For MOZAIC/IAGOS observations in the Ex-UTLS Cohen et al. (2018) report, e.g., for springtime lowermost  
 stratosphere conditions values of  $[O_3]_{\text{med}} = 400$  ppbv and  $[O_3]_{\text{P95}} = 600$  ppbv, resulting in  $m = 0.36$ , and for  
 tropopause layer conditions values of  $[O_3]_{\text{med}} = 110$  ppbv,  $[O_3]_{\text{P95}} = 200$  ppbv, and  $m = 0.57$ ; note that P95 refers  
 590 here to the 95 percentile value of the analysed data ensemble, [as taken from Cohen et al. \(2018\)](#). Deeper into the  
 stratosphere beyond the reach of MOZAIC/IAGOS aircraft, the value of  $m$  approaches  $m = 0.0$ . Thus, similar to  
 the troposphericity parameter defined by Cirisan et al. (2013) from trajectory analyses, a value of  $m = 0$  indicates  
 that an air parcel contains only stratospheric air, while  $m = 1$  is fully tropospheric. Defining the troposphericity  
 as described here, we connect the troposphericity of an air mass to the observed variability of the  $O_3$  VMR.



**Figure 11.** Vertical distribution of the troposphericity parameter  $m$  for ISSR and non-ISSR air masses with respect to the thermal (a) and dynamical (b) tropopause.

600 The analysis of troposphericity of the seven investigated layers with respect to the 99 percentile and median  $O_3$   
 VMRs is presented in Figure 11. With respect to the thermal as well as to the dynamical tropopause, the layers  
 up to the tropopause layer are characterised by almost similar values of  $m > 0.80$  for ISSR and  $m > 0.75$  for non-  
 ISSR air masses. The first layer above the thermal tropopause, however, shows a clear difference between ISSR  
 ( $m = 0.67$ ) and non-ISSR ( $m = 0.44$ ) with respect to the thermal tropopause, but similar values of  $m = 0.67 - 0.69$   
 605 for ISSR and non-ISSR with respect to the dynamical tropopause.

[Recalling the structure of the ExTL with the 2 PVU dynamical tropopause at its lower bound separating the stratosphere from the troposphere, and centred on the thermal tropopause, we find that on top of the ExTL. With respect to the thermal tropopause, we find a distinct difference in the mixing behaviour. Above the transport](#)

~~barrier formed by the thermal tropopause, non-ISSR air masses show a clear stratospheric signature, while. In contrast, ISSR air masses just above the tropopause are still strongly influenced by mixing and carry a of significant tropospheric nature-fingerprint compared to the non-ISSR air masses. Above the Referring to the dynamical tropopause and thus inside the ExTL, the influence of mixing increases gradually for both ISSR and non-ISSR air masses and the difference in troposphericity is much less pronounced than in the case of the thermal tropopause near the top of the ExTL.~~

~~Concluding, for both tropopause definitions used in our analysis, the troposphericity values indicate continuing tropospheric influence for ISSR inside and above the respective tropopause layer. For the non-ISSR, however, the stratospheric nature is clearly visible above the thermal tropopause whereas above the dynamical tropopause the air masses show still a relevant tropospheric influence. All investigated properties demonstrate the efficiency of the thermal tropopause as a vertical transport barrier, while the air masses between the dynamical and the thermal tropopauses are clearly shaped by mixing with tropospheric air.~~

### 3.4 ISSR fraction and cirrus cloud occurrence

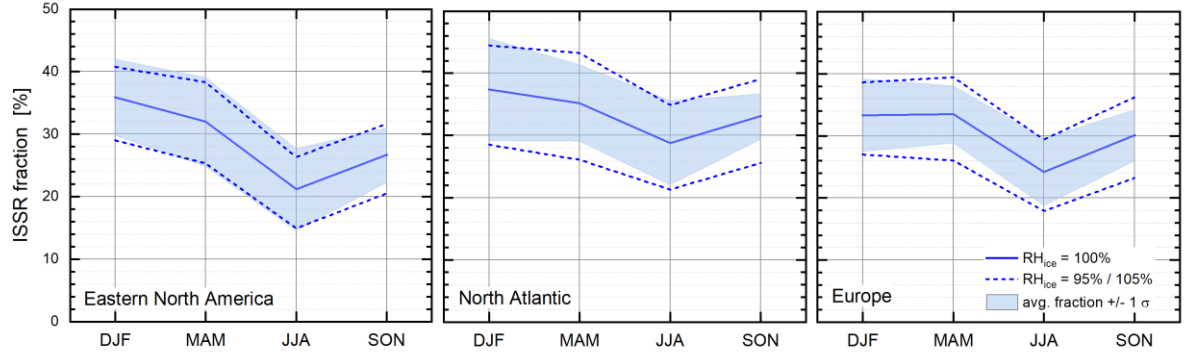
Ice-supersaturation in the atmosphere is a prerequisite for the formation of cirrus clouds, and the degree of supersaturation, mostly driven by atmospheric dynamics, determines the mechanism by which ice particles form (e.g., Kärcher et al., 2014; Krämer et al., 2016; Heymsfield et al., 2017). ~~Furthermore, the analysis of a large set of combined observation of  $RH_{ice}$  and ice crystal number concentration  $N_{ice}$  during a series of research flights (approx. 68000 observations of ice-supersaturation; Krämer et al., 2016) demonstrated, that approx. 80 % of the observed ice-supersaturation events are associated with in-cloud conditions. On the other hand,  $RH_{ice}$  probability distribution functions inside cirrus clouds are characterised by most probable values at or slightly above ice-saturation at  $RH_{ice} = 100\%$  (Krämer et al., 2009; Diao et al., 2014; Diao et al., 2015; Petzold et al., 2017) which means that cirrus clouds exist to a considerable fraction also in ice-subaturated air masses, depending on their state of life. Finally, Iceice-supersaturation can also occur in cloud-free air masses, but the fraction of ice-supersaturated air in clear sky conditions is largely unknown. However, these cloud-free ISSR are of high importance for the formation of persistent contrails and thus for the climate impact of aviation (Irvine and Shine, 2015; Kärcher, 2018).~~

~~Driven-Motivated~~ by the high importance of ISSR for cirrus formation and existence and also for the formation and persistence of contrails, we converted the vertically resolved observations of ISSR fractions into an annual cycle of ISSR occurrence for the three target regions. The seasonal-mean occurrence probabilities were analysed for  $RH_{ice}$  values of 95%, 100% and 105%, based on the sensor precision of 5%  $RH_{ice}$ . The resulting annual cycles for the top two UT layers, situated just below the thermal tropopause layer are shown in Figure 12. The range bound by the probabilities of occurrence for  $RH_{ice} = 95\%$  and 105% defines the uncertainty of our analysis. Additionally, we analysed the interannual variability of ISSR occurrence from the standard deviation of the mean ISSR occurrence probability for  $RH_{ice} = 100\%$ . The respective variability range is shown as blue-shaded areas in Figure 12. ~~It becomes clearly visible, that the sensor uncertainty and the interannual variability of the ISSR occurrence probability cover similar ranges.~~

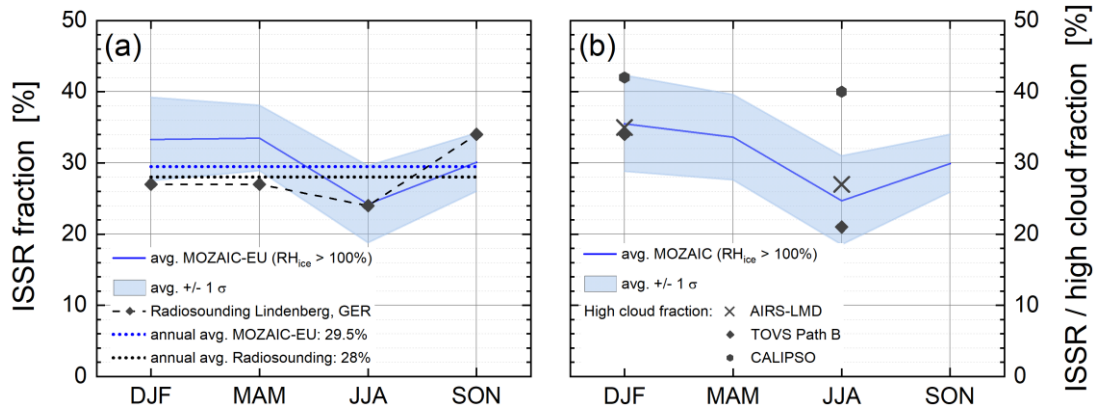
For all regions, ISSR occurrence probabilities are highest in winter/spring and lowest in summer, while the absolute values particularly in summer are considerably different. The probability for finding ice-supersaturated air masses during summer is 20% over the Eastern North America regions, but 30% over the North Atlantic, with Europe showing values in the range between.



To the present, there is only very limited in-situ information available about the occurrence probability of ice-supersaturated air masses in the upper troposphere in general and about their seasonality in particular. One source for in-situ information stems from radiosonde observations conducted by the German Weather Service over the observatory Lindenberg in Germany (Spichtinger et al., 2003a).



**Figure 12.** Annual cycles of ISSR occurrence shown as occurrence probability for  $RH_{ice} > 100\%$ , for the regions Eastern North America, North Atlantic and Europe; considered years are 1995 to 2010, with shaded areas representing probabilities for the average value (thick lines)  $\pm 1\sigma$ , and the short-dashed lines representing average fractions for  $RH_{ice} = 95\%$  and  $105\%$ , respectively; calculations were conducted for the two UT layers positioned closest to the thermal tropopause.



**Figure 13.** a: Seasonal cycle of ISSR occurrence probability, i.e.  $p(RH_{ice} > 100\%)$ , averaged over Europe for the years 1995 to 2010 for the two UT layers positioned closest to the thermal tropopause; symbols represent the annual cycle of the Lindenberg sounding (2000 – 2001) from Spichtinger et al. (2003a); b: Seasonal cycle of ISSR occurrence probability, as  $p(RH_{ice} > 100\%)$  averaged over the Northern Mid-Latitudes from East North America to Europe for the period 1995 to 2010; symbols represent high cloud fractions from the satellite cloud climatology by Stubenrauch et al. (2010) for northern mid-latitudes and years 2003 to 2008 for AIRS-LMD, 1987 to 1995 for TOVS Path B and 2006 to 2007 for CALIPSO.

Figure 13a shows the average annual cycles of ISSR occurrence frequency from 15 years of MOZAIC observations over Europe and from 15 months of radiosonde observations over Lindenberg published by Spichtinger et al. (2003a). The single annual 15-months cycle from the radio soundings fits well into the

~~covered by the~~ 15 years climatology of ISSR occurrence from MOZAIC, ~~but contributes only a snapshot compared to the 15-years' time series. More quantitatively~~Based on the 15 months of observation, the authors report a mean frequency of occurrence of ice-supersaturation layers over Lindenberg of 28%, whereas the annual  
675 cycle of ISSR occurrence from our 15 years of MOZAIC observations over Europe yields a mean value of 29.5% with a range from 35% ( $RH_{ice} = 95\%$ ) to 23% ( $RH_{ice} = 105\%$ ).

Another source of data, but for the occurrence frequency of cirrus clouds originates from long-term analyses of satellite observations (Stubenrauch et al., 2010; Stubenrauch et al., 2013). In their 6-year climatology Stubenrauch et al. (2010) report cirrus cloud coverage fractions for northern mid-latitudes of 35% in January and  
680 27% in July from AIRS-LMD (2003 to 2008), and respective fractions of 34% and 21% from TOVS – Path B (1987 to 1995), and 42% and 40% from CALIPSO (2006 to 2007). The compilation of our annual cycle of ISSR occurrence and the respective observations from space-borne sensors is shown in Figure 13b. The agreement of the observations of ISSR occurrence from the very different sources is remarkably good, with the exception of CALIPSO observations which provide higher values. According to Stubenrauch et al. (2010), the high cloud  
685 fraction of CALIPSO is about 10% larger than respective values of CALIPSO for clouds excluding subvisible cirrus. Therefore, the difference between high cloud fractions from CALIPSO and from the other instruments shown in Figure 13 can be attributed to instrument sensitivities.

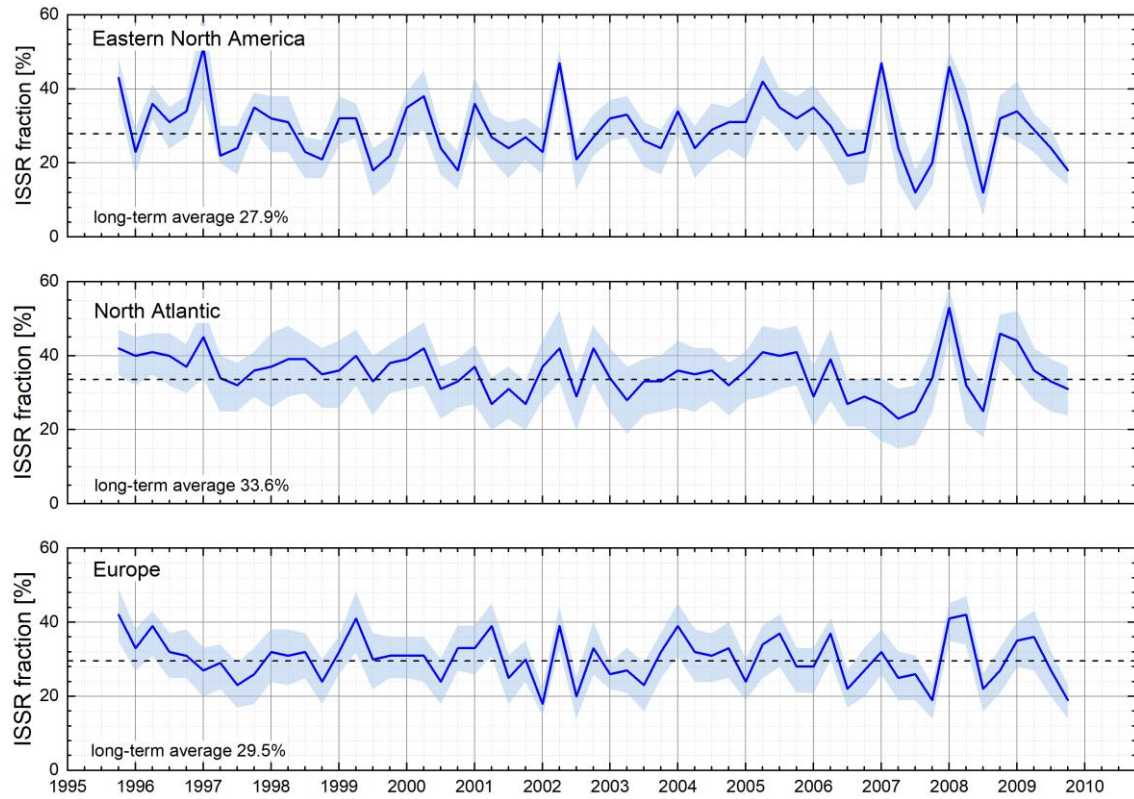
~~It has to be noted, however, that CALIPSO also detects subvisible cirrus clouds which are below the detection limit of the other instruments.~~

The ~~close match~~good agreement between MOZAIC in-situ observations of  ~~$RH_{ice}$ -ISSR occurrence~~ with the high-  
690 cloud fraction from satellite instruments encourages further detailed studies on this matter,  
~~corresponds to the finding from other studies that by far the largest part of ISSRs occurs inside cirrus clouds. We find from the analysis of a large set of combined observation of  $RH_{ice}$  and ice crystal number concentration  $N_{ice}$  during a series of research flights, that approx. 80 % of the observed ice supersaturation events are associated~~  
695 ~~with in-cloud conditions.~~ First exemplary analyses of simultaneous observations of  $RH_{ice}$  and  $N_{ice}$  which are now possible within the ongoing IAGOS programme also already indicate a strong correlation of high  $RH_{ice}$  values with its occurrence inside cirrus clouds (Petzold et al., 2017). ~~Further studies on this topic will be launched as soon as the full data set of combined observations of  $RH_{ice}$  and  $N_{ice}$  from IAGOS flights since 2011 is validated.~~

### 3.5 Trend analysis

700 Finally, we analysed the 15-years records of the validated MOZAIC  $RH_{ice}$  observations and the resulting fraction of ISSR observations for the three regions Eastern North America, North Atlantic and Europe for potential trends. The bases of our analyses were the seasonally averaged observations in the uppermost tropospheric layer (UT) with respect to the thermal tropopause, and the respective average seasonal cycles depicted in Figure 12. The resulting time series are shown in Figure 14. The seasonality of ISSR occurrence is clearly visible for each  
705 region, but with considerable interannual variability. Similar to Figure 12, the shaded regions represent the average fractions for  $RH_{ice} = 95\%$  and  $105\%$ , respectively, and indicate thus the uncertainty resulting from the instrument precision of  $RH_{ice} = 5\%$ . For none of the regions, we find significant trends in ISSR occurrence. Therefore, the distribution of  $RH_{ice}$  in the uppermost troposphere close to the tropopause layer and the resulting occurrence of ice-supersaturation seem to be stable over the investigated time period from 1995 to 2010.

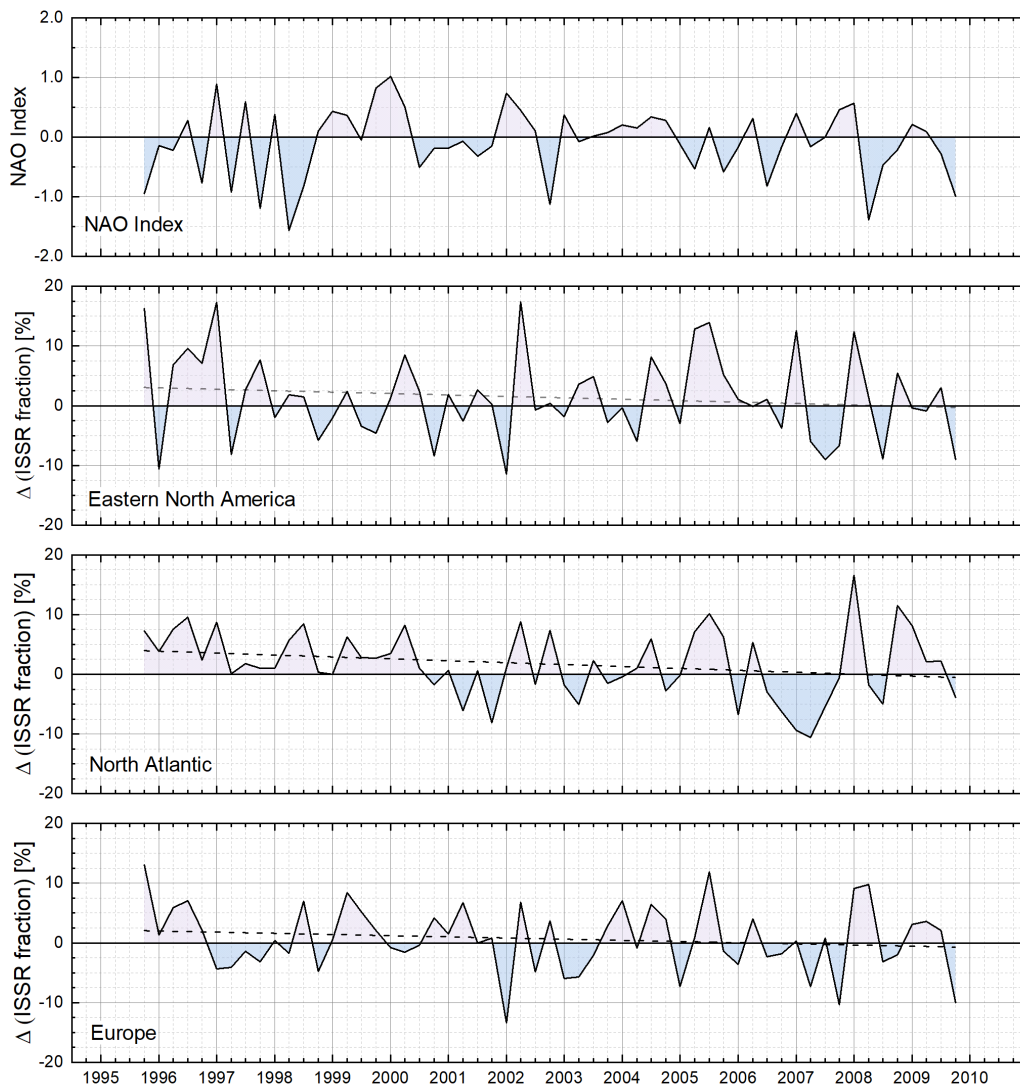
710 In order to get a clearer understanding of the reasons for the interannual variability, we further analysed the de-  
 seasonalised time series of the ISSR fractions by calculating the difference between each seasonal value of the  
 ISSR fraction and the 15-years average seasonal cycle (see Figure 12).



**Figure 14.** Time series of ISSR fraction (probability of occurrence) for latitudes 40°N to 60°N and for the  
 715 regions (from top to bottom) Eastern North America (105°W to 65°W), North Atlantic (65°W to 5°W) and  
 Europe (5°W to 30°E) for the top UT layer, situated just below the tropopause layer; with the solid lines  
 representing probabilities for the average value for  $RH_{ice} = 100\%$  and the shaded areas representing average  
 fractions for  $RH_{ice} = 95\%$  and  $105\%$ , respectively. Long-term average values for  $RH_{ice} = 100\%$  are added in the  
 panels.

720





**Figure 15.** De-seasonalised time series of ISSR fraction (probability of occurrence) for latitudes 40°N to 60°N and for the regions (from top to bottom) Eastern North America (105°W to 65°W), North Atlantic (65°W to 5°W) and Europe (5°W to 30°E).

725

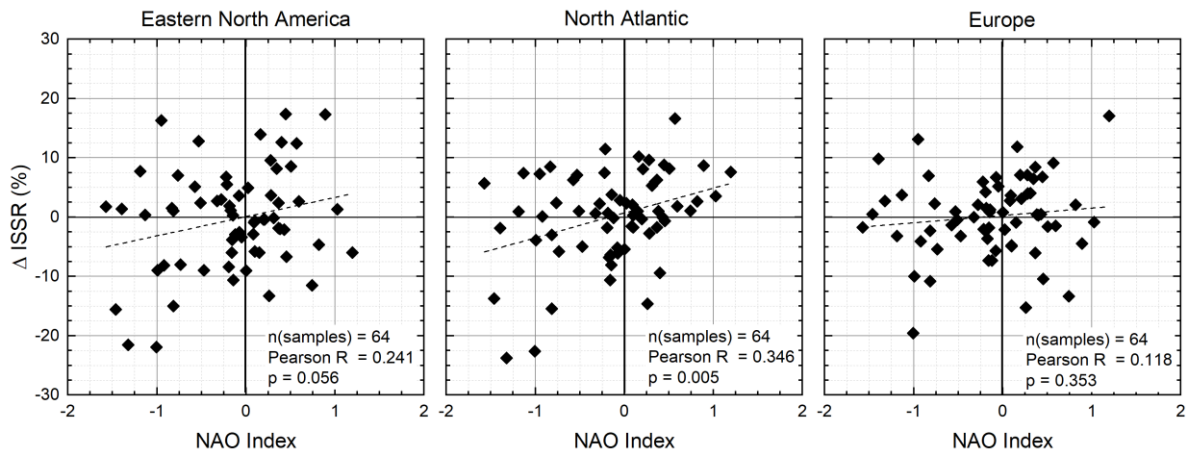
The de-seasonalised time-series thus show positive and negative deviations from the long-term seasonal average values. The resulting time series are presented in Figure 15. As for the time series of ISSR occurrence, we performed a trend analysis and added the obtained trend lines to Figure 15. Respective decadal slopes are  $-1.95\% \pm 1.77\%$  for Eastern North America,  $-3.21\% \pm 1.78\%$  for the North Atlantic, and  $-2.39\% \pm 2.29\%$  for Europe and indicated uncertainties of the determined slopes refer to one standard deviation. Thus none of the slopes differs significantly from zero, and similar to the time series of ISSR occurrence, we do not observe significant trends for the seasonal deviation of ISSR occurrence from the long-term average for the three target regions.

730

One potential weather phenomenon driving the deviation of seasonal ISSR occurrence from the long-term average in the investigated region is the North Atlantic Oscillation (NAO). The NAO index describes the deviation of the pressure difference between the Iceland low and the Azores high pressure systems from the

735

long-term average value. As an example, a positive value of the NAO index indicates that  $\Delta p$  (Iceland L to Azores H)



**Figure 16.** Cluster-Correlation analysis with respect to the correlation of signs between NAO index and deviation of ISSR occurrence from the long-term average ( $\Delta$  ISSR) for the target regions; numbers indicate the results from the correlation analysis with respect to number of samples n, Pearson R and significance level p; black symbols and grey shaded clusters indicate the same sign for NAO index and  $\Delta$  ISSR, red symbols and red shaded clusters indicate opposite signs.

is larger than on average. This larger pressure difference causes stronger westerly winds and thereby more active storm tracks over the North Atlantic ~~which brings warmer and more moist air to Europe~~. Under such conditions we would expect a higher probability of ice-supersaturation in the uppermost troposphere due to more frequent warm conveyor belts that can induce the formation of ISSRs in the upper troposphere (Spichtinger et al., 2005). Such a positive correlation between NAO and cirrus cloud cover is reported from an analysis of cirrus cloud cover data from the International Satellite Cloud Climatology Project and relative humidity data from ECMWF/ERA40 by Eleftheratos et al. (2007).

To investigate this potential link, we added the seasonally averaged NAO index to Figure 15 (top panel). Since there is no immediate evidence given for a link between the NAO index and the deviation of ISSR occurrence from the long-term average ( $\Delta$  ISSR), we further searched for a potential link of signs in the sense that positive and negative NAO index values are associated ~~to~~ with positive and negative deviations of ISSR occurrence from the long-term average, respectively. The results of this cluster-cross-correlation analysis are presented in

**Figure 16.**

For the regions Eastern North America and Europe the correlation between NAO index and  $\Delta$  ISSR is not statistically significant. For the North Atlantic however, the results of the cross-correlation analysis indicate statistical significance at a level of 99%. probability of cross-correlated signs between NAO index and  $\Delta$  ISSR fraction is 52% ( $p = 0.52$ ) whereas the probability for correlated signs is 48% ( $p = 0.48$ ). The probabilities are almost equal for this region and from that result we conclude that there is no link between NAO index and  $\Delta$  ISSR fraction over the eastern part of North America. In contrast, the results are different for the regions North Atlantic and Europe, both of which show correlated signs between NAO index and  $\Delta$  ISSR in 61% and 58% of the analysed seasons, respectively. For these regions, we consider the correlation of signs statistically significant.

The obtained correlation of signs is in line with the observation that the occurrence of ice-supersaturation is well correlated with the storm track activity (Spichtinger et al., 2003b; Gettelman et al., 2006; Lamquin et al., 2012).

#### 4. Summary and Conclusions

The European Research Infrastructure IAGOS (from 2011) and its predecessor programme MOZAC (1994 - 2010) perform global-scale routine in-situ observations of relative humidity with respect to ice ( $RH_{ice}$ ) by using instrumented passenger aircraft. The validated  $RH_{ice}$  data set from the MOZAIC period between 1995 and 2010 was analysed for latitudes 40 °N to 60 °N and for the regions Eastern North America (105 °W to 65 °W), North Atlantic (65 °W to 5 °W) and Europe (5 °W to 30 °E) to study the occurrence of ice-supersaturated regions (ISSR) in the uppermost troposphere and tropopause layers. Determined seasonal cycles agree very well with observations of ISSR occurrence from radio soundings (Spichtinger et al., 2003a) and from satellite observations (Spichtinger et al., 2003b; Lamquin et al., 2012).

The high vertical resolution of the MOZAIC  $RH_{ice}$  data set with 30 hPa layer thickness allows the determination of the vertical position of the ice-supersaturated air masses with respect to the thermal tropopause. It occurs that the fraction of ice-supersaturated regions is largest for the atmospheric layers of 60 hPa thickness, directly below the thermal tropopause.

Comparing the ISSR fraction from MOZAIC in-situ observations with the high-cloud fraction from satellite instruments (Stubenrauch et al., 2010) yields remarkably close agreement between the two different observations and supports the interpretation that cirrus clouds exist to a considerable fraction also in ice-subaturated air masses, depending on their state of life. by far the largest part of the ice-supersaturation occurs inside cirrus clouds. This interpretation is also supported by first exemplary analyses of simultaneous observations of  $RH_{ice}$  and ice crystal number density  $N_{ice}$  from the ongoing IAGOS programme (Petzold et al., 2017). In addition, the close agreement between satellite-based observations of ice cloud occurrence and the MOZAIC/IAGOS in-situ observations of ice-supersaturation demonstrate the unique contribution, MOZAIC and today IAGOS long-term observations can make to this scientific area, in particular with the detailed seasonality of ISSR occurrence over different regions. Future work will combine  $RH_{ice}$  and  $N_{ice}$  observations which are now available from IAGOS and link them to AIRS time series.

The finding that ice-supersaturated air is generally colder and more humid associated with higher  $RH_{ice}$  and – in case of observation inside or above the tropopause layer - carries less ozone than the surrounding air masses is in close agreement with reported results for temperature and relative humidity. However, we were also able to use ozone as a tracer for stratospheric air and calculate the troposphericity of ice-supersaturated and subsaturated air masses. The analysis yields a significant impact of tropospheric air even on ISSR observed above the thermal tropopause. The thermodynamic features together with the increased troposphericity indicate vertical mixing in the vicinity of the tropopause layer as one important formation process of ice-supersaturation. Future work in this direction will be conducted, once the full IAGOS data set on  $RH_{ice}$ , ozone and ice clouds is validated and available.

Over the investigated period of 15 years, no significant trends are observed, neither for the occurrence of ISSR nor for the deviation of seasonal ISSR occurrence probabilities from the long-term average. This statement is valid for all three investigated regions. Yet, we identify a significant correlations of signs between the NAO index and the deviation of seasonal ISSR occurrence probabilities from the long-term average for the North Atlantic and Europe regions, whereas no such correlation was found for the Eastern North America and

Europe region. The resulting interpretation is that a positive NAO index correlates with increased occurrence of ISSR (positive deviation from the long-term average). This interpretation is in agreement with the understanding that a positive NAO index leads to an increased storm track activity which then may induce more frequent formation of ISSRs in the upper troposphere generates more frequently ISSR.

Finally, in a concomitant study by Reutter et al. (2020) MOZAIC RH<sub>ice</sub> observations have been compared to ECMWF ERA-Interim data and significant deviations are reported for ice-supersaturated conditions, both in number and strength of supersaturation. The high quality and very good resolution of MOZAIC and later IAGOS RH<sub>ice</sub> observations will certainly help to further improve the representation of ice-supersaturation in ERA 5 as well as in numerical weather and climate forecasting models.

### Author contributions

AP designed the study and prepared the manuscript, with contributions from all co-authors; PN, SR, MR, and HGJS performed the quality control and analysis of MOZAIC/IAGOS water vapour data; FB provided the thermal tropopause pressure levels and performed the quality control and analysis of temperature data; MK and NS contributed the analysis of the research aircraft data. PNeD performed the quality control and analysis of ozone data; AW and PS contributed to the interpretation of the study results.

### Competing interests

The authors declare that they have no conflict of interest.

### Data Availability

The IAGOS data are available through the IAGOS data portal at <https://doi.org/10.25326/20>. The IAGOS time series data set used for this analysis is referenced at https://doi.org/10.25326/06.

We used the following data versions for our analyses:

Version 1.0 of IAGOS air\_temp and air\_stag\_temp data, based on the method described in Helten et al. (1998).

Version 3.0 of IAGOS RHL, RHI and H<sub>2</sub>O\_gas data, based on the calibration techniques and data inversion algorithms published in Helten et al. (1998). In addition, version 3.0 has implemented the in-flight calibration technique adapted from Smit et al. (2008), which adjusts for an offset drift of the MCH sensor during a flight period.

### Acknowledgements

Parts of this study were funded by the German Ministry for Education and Research (BMBF) under Grant No. 01LK1301A as part of the joint research programme IAGOS Germany. MOZAIC/IAGOS data are created with support from the European Commission, national agencies in Germany (BMBF), France (MESR), and the UK (NERC), and the IAGOS member institutions (<http://www.iagos.org/partners>). The participating airlines (Deutsche Lufthansa, Air France, Austrian, China Airlines, Iberia, Cathay Pacific, Air Namibia, Sabena) supported IAGOS by carrying the measurement equipment free of charge since 1994. The data are available at <https://doi.org/10.25326/20> thanks to additional support from AERIS. MK thanks JGU Mainz for support as a GfK fellow.

## References

- Aaltonen, V., Lihavainen, H., Kerminen, V. M., Komppula, M., Hatakka, J., Eneroth, K., Kulmala, M., and Viisanen, Y.: Measurements of optical properties of atmospheric aerosols in Northern Finland, *Atmos. Chem. Phys.*, 6, 1155-1164, 2006.
- 850 Anderson, J. G., Wilmouth, D. M., Smith, J. B., and Sayres, D. S.: UV Dosage Levels in Summer: Increased Risk of Ozone Loss from Convectively Injected Water Vapor, *Science*, 337, 835-839, doi: 10.1126/science.1222978, 2012.
- Anderson, J. G., Weisenstein, D. K., Bowman, K. P., Homeyer, C. R., Smith, J. B., Wilmouth, D. M., Sayres, D. S., Klobas, J. E., Leroy, S. S., Dykema, J. A., and Wofsy, S. C.: Stratospheric ozone over the United States in summer linked to observations of convection and temperature via chlorine and bromine catalysis, *Proc. Natl. Acad. Sci. U. S. A.*, 114, E4905-E4913, doi: 10.1073/pnas.1619318114, 2017.
- 855 Berkes, F., Neis, P., Schultz, M. G., Bundke, U., Rohs, S., Smit, H. G. J., Wahner, A., Konopka, P., Boulanger, D., Nédélec, P., Thouret, V., and Petzold, A.: In situ temperature measurements in the upper troposphere and lowermost stratosphere from 2 decades of IAGOS long-term routine observation, *Atmos. Chem. Phys.*, 17, 12495-12508, doi: 10.5194/acp-17-12495-2017, 2017.
- 860 Bock, L., and Burkhardt, U.: Contrail cirrus radiative forcing for future air traffic, *Atmos. Chem. Phys.*, 19, 8163-8174, doi: 10.5194/acp-19-8163-2019, 2019.
- Bodeker, G. E., Bojinski, S., Cimini, D., Dirksen, R. J., Haeffelin, M., Hannigan, J. W., Hurst, D. F., Leblanc, T., Madonna, F., Maturilli, M., Mikalsen, A. C., Philipona, R., Reale, T., Seidel, D. J., Tan, D. G. H., Thorne, P. W., Vomel, H., and Wang, J.: Reference upper air observations for climate: From concept to reality, *Bull. Amer. Meteorol. Soc.*, 97, 123-135, doi: 10.1175/bams-d-14-00072.1, 2016.
- 865 Boucher, O., Randall, D., Artaxo, P., Bretherton, C., Feingold, G., Forster, P., Kerminen, V.-M., Kondo, Y., Liao, H., Lohmann, U., Rasch, P., Satheesh, S. K., Sherwood, S., Stevens, B., and Zhang, X. Y.: Clouds and Aerosols, in: *Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change*, edited by: Stocker, T. F., D. Qin, Plattner, G.-K., Tignor, M., Allen, S. K., Boschung, J., Nauels, A., Xia, Y., Bex, V., and Midgley, P. M., Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, 2013.
- 870 Brenninkmeijer, C. A. M., Crutzen, P. J., Fischer, H., Gusten, H., Hans, W., Heinrich, G., Heintzenberg, J., Hermann, M., Immelmann, T., Kersting, D., Maiss, M., Nolle, M., Pitscheider, A., Pohlkamp, H., Scharffe, D., Specht, K., and Wiedensohler, A.: CARIBIC - Civil aircraft for global measurement of trace gases and aerosols in the tropopause region, *J. Ocean. Atmos. Technol.*, 16, 1373-1383, doi: 10.1175/1520-0426(1999)016<1373:cacafgm>2.0.co;2, 1999.
- 875 Brenninkmeijer, C. A. M., Crutzen, P., Boumard, F., Dauer, T., Dix, B., Ebinghaus, R., Filippi, D., Fischer, H., Franke, H., Friess, U., Heintzenberg, J., Helleis, F., Hermann, M., Kock, H. H., Koepfel, C., Lelieveld, J., Leuenberger, M., Martinsson, B. G., Miemczyk, S., Moret, H. P., Nguyen, H. N., Nyfeler, P., Oram, D., O'Sullivan, D., Penkett, S., Platt, U., Pupek, M., Ramonet, M., Randa, B., Reichelt, M., Rhee, T. S., Rohwer, J., Rosenfeld, K., Scharffe, D., Schlager, H., Schumann, U., Slemr, F., Sprung, D., Stock, P., Thaler, R., Valentino, F., van Velthoven, P., Waibel, A., Wandel, A., Waschitschek, K., Wiedensohler, A., Xueref-Remy, I., Zahn, A., Zech, U., and Ziereis, H.: Civil Aircraft for the regular investigation of the atmosphere based on an instrumented container: The new CARIBIC system, *Atmos. Chem. Phys.*, 7, 4953-4976, 2007.
- 880 Buchholz, B., Kuehnreich, B., Smit, H. G. J., and Ebert, V.: Validation of an extractive, airborne, compact TDL spectrometer for atmospheric humidity sensing by blind intercomparison, *Appl. Phys. B*, 110, 249-262, doi: 10.1007/s00340-012-5143-1, 2013.
- 885 Burkhardt, U., Kärcher, B., Ponater, M., Gierens, K., and Gettelman, A.: Contrail cirrus supporting areas in model and observations, *Geophys. Res. Lett.*, 35, doi: 10.1029/2008gl034056, 2008.
- Burkhardt, U., and Kärcher, B.: Global radiative forcing from contrail cirrus, *Nature*, 1, 54-58, doi: doi:10.1038/NCLIMATE1068, 2011.
- 890 Chen, T., Rossow, W. B., and Zhang, Y. C.: Radiative effects of cloud-type variations, *J. Clim.*, 13, 264-286, doi: 10.1175/1520-0442(2000)013<0264:Reoctv>2.0.Co;2, 2000.
- Cirisan, A., Spichtinger, P., Luo, B. P., Weisenstein, D. K., Wernli, H., Lohmann, U., and Peter, T.: Microphysical and radiative changes in cirrus clouds by geoengineering the stratosphere, *J. Geophys. Res.-Atmos.*, 118, 4533-4548, doi: 10.1002/jgrd.50388, 2013.
- 895 Cohen, Y., Petetin, H., Thouret, V., Maréchal, V., Josse, B., Clark, H., Sauvage, B., Fontaine, A., Athier, G., Blot, R., Boulanger, D., Cousin, J. M., and Nédélec, P.: Climatology and long-term evolution of ozone and carbon monoxide in the upper troposphere-lower stratosphere (UTLS) at northern midlatitudes, as seen by IAGOS from 1995 to 2013, *Atmos. Chem. Phys.*, 18, 5415-5453, doi: 10.5194/acp-18-5415-2018, 2018.
- 900 Dee, D. P., Uppala, S. M., Simmons, A. J., Berrisford, P., Poli, P., Kobayashi, S., Andrae, U., Balmaseda, M. A., Balsamo, G., Bauer, P., Bechtold, P., Beljaars, A. C. M., van de Berg, L., Bidlot, J., Bormann, N., Delsol, C., Dragani, R., Fuentes, M., Geer, A. J., Haimberger, L., Healy, S. B., Hersbach, H., Holm, E. V., Isaksen, I., Kallberg, P., Kohler, M., Matricardi, M., McNally, A. P., Monge-Sanz, B. M., Morcrette, J. J., Park, B. K., Peubey, C., de Rosnay, P., Tavolato, C., Thepaut, J. N., and Vitart, F.: The ERA-Interim reanalysis: configuration and performance of the data assimilation system, *Q. J. R. Meteorol. Soc.*, 137, 553-597, doi: 10.1002/qj.828, 2011.
- 905 Diao, M., Zondlo, M. A., Heymsfield, A. J., Avallone, L. M., Paige, M. E., Beaton, S. P., Campos, T., and Rogers, D. C.: Cloud-scale ice-supersaturated regions spatially correlate with high water vapor heterogeneities, *Atmos. Chem. Phys.*, 14, 2639-2656, doi: 10.5194/acp-14-2639-2014, 2014.
- Diao, M., Jensen, J. B., Pan, L. L., Homeyer, C. R., Honomichl, S., Bresch, J. F., and Bansemer, A.: Distributions of ice supersaturation and ice crystals from airborne observations in relation to upper tropospheric dynamical boundaries, *J. Geophys. Res.-Atmos.*, 120, 5101-5121, doi: 10.1002/2015jd023139, 2015.
- 910 Dyroff, C., Zahn, A., Christner, E., Forbes, R., Tompkins, A. M., and van Velthoven, P. F. J.: Comparison of ECMWF analysis and forecast humidity data with CARIBIC upper troposphere and lower stratosphere observations, *Q. J. R. Meteorol. Soc.*, doi: 10.1002/qj.2400, 2014.

- 915 Eleftheratos, K., Zerefos, C. S., Zanis, P., Balis, D. S., Tselioudis, G., Gierens, K., and Sausen, R.: A study on natural and manmade global interannual fluctuations of cirrus cloud cover for the period 1984-2004, *Atmos. Chem. Phys.*, 7, 2631-2642, doi: 10.5194/acp-7-2631-2007, 2007.
- Gottelman, A., Fetzer, E. J., Eldering, A., and Irion, F. W.: The global distribution of supersaturation in the upper troposphere from the Atmospheric Infrared Sounder, *J. Climate*, 19, 6089-6103, doi: 10.1175/jcli3955.1, 2006.
- 920 Gottelman, A., Hoor, P., Pan, L. L., Randel, W. J., Hegglin, M. I., and Birner, T.: The extratropical upper troposphere and lower stratosphere, *Rev. Geophys.*, 49, RG3003, doi: 10.1029/2011rg000355, 2011.
- Gottelman, A., Liu, X., Barahona, D., Lohmann, U., and Chen, C.: Climate impacts of ice nucleation, *J. Geophys. Res.-Atmos.*, 117, D20201, doi: 10.1029/2012jd017950, 2012.
- Gierens, K., Schumann, U., Helten, M., Smit, H., and Marenco, A.: A distribution law for relative humidity in the upper troposphere and lower stratosphere derived from three years of MOZAIC measurements, *Ann. Geophys.*, 17, 1218-1226, doi: 925 10.1007/s005850050846, 1999.
- Gierens, K., Schumann, U., Helten, M., Smit, H., and Wang, P. H.: Ice-supersaturated regions and subvisible cirrus in the northern midlatitude upper troposphere, *J. Geophys. Res.*, 105, 22743-22753, doi: 10.1029/2000jd900341, 2000.
- Gierens, K., and Spichtinger, P.: On the size distribution of ice-supersaturated regions in the upper troposphere and lowermost stratosphere, *Ann. Geophys.*, 18, 499-504, doi: 10.1007/s005850050907, 2000.
- 930 Gierens, K., and Brinkop, S.: Dynamical characteristics of ice supersaturated regions, *Atmos. Chem. Phys.*, 12, 11933-11942, doi: 10.5194/acp-12-11933-2012, 2012.
- Gierens, K., Eleftheratos, K., and Shi, L.: Technical Note: 30 years of HIRS data of upper tropospheric humidity, *Atmos. Chem. Phys.*, 14, 7533-7541, doi: 10.5194/acp-14-7533-2014, 2014.
- 935 Helten, M., Smit, H. G. J., Sträter, W., Kley, D., Nédélec, P., Zöger, M., and Busen, R.: Calibration and performance of automatic compact instrumentation for the measurement of relative humidity from passenger aircraft, *J. Geophys. Res.*, 103, 25643-25652, doi: 10.1029/98jd00536, 1998.
- Helten, M., Smit, H. G. J., Kley, D., Ovarlez, J., Schlager, H., Baumann, R., Schumann, U., Nédélec, P., and Marenco, A.: In-flight comparison of MOZAIC and POLINAT water vapor measurements, *J. Geophys. Res.*, 104, 26087-26096, doi: 10.1029/1999jd900315, 1999.
- 940 Heymsfield, A. J., Krämer, M., Luebke, A., Brown, P., Cziczko, D. J., Franklin, C., Lawson, P., Lohmann, U., McFarquhar, G., Ulanowski, Z., and Tricht, K. V.: Cirrus Clouds, *Meteor. Monogr.*, 58, 2.1-2.26, doi: 10.1175/amsmonographs-d-16-0010.1, 2017.
- Hoor, P., Gurk, C., Brunner, D., Hegglin, M. I., Wernli, H., and Fischer, H.: Seasonality and extent of extratropical TST derived from in-situ CO measurements during SPURT, *Atmos. Chem. Phys.*, 4, 1427-1442, doi: 10.5194/acp-4-1427-2004, 945 2004.
- Hoor, P., Wernli, H., Hegglin, M. I., and Boenisch, H.: Transport timescales and tracer properties in the extratropical UTLS, 10, 7929-7944, doi: 10.5194/acp-10-7929-2010, 2010.
- Hoose, C., and Möhler, O.: Heterogeneous ice nucleation on atmospheric aerosols: a review of results from laboratory experiments, *Atmos. Chem. Phys.*, 12, 9817-9854, doi: 10.5194/acp-12-9817-2012, 2012.
- 950 Irvine, E. A., Hoskins, B. J., and Shine, K. P.: A Lagrangian analysis of ice-supersaturated air over the North Atlantic, *J. Geophys. Res.*, 119, 90-100, doi: 10.1002/2013jd020251, 2014.
- Irvine, E. A., and Shine, K. P.: Ice supersaturation and the potential for contrail formation in a changing climate, *Earth Syst. Dynam.*, 6, 555-568, doi: 10.5194/esd-6-555-2015, 2015.
- Ivanova, A. R.: The tropopause: Variety of definitions and modern approaches to identification, *Russ. Meteorol. Hydrol.*, 38, 808-817, doi: 10.3103/s1068373913120029, 2013.
- 955 Jöckel, P., Tost, H., Pozzer, A., Kunze, M., Kirner, O., Brenninkmeijer, C. A. M., Brinkop, S., Cai, D. S., Dyroff, C., Eckstein, J., Frank, F., Garny, H., Gottschaldt, K. D., Graf, P., Grewe, V., Kerkweg, A., Kern, B., Matthes, S., Mertens, M., Meul, S., Neumaier, M., Nutzel, M., Oberlander-Hayn, S., Ruhnke, R., Runde, T., Sander, R., Scharffe, D., and Zahn, A.: Earth System Chemistry integrated Modelling (ESCiMo) with the Modular Earth Submodel System (MESSy) version 2.51, *Geosci. Model Dev.*, 9, 1153-1200, doi: 10.5194/gmd-9-1153-2016, 2016.
- 960 Kärcher, B., and Lohmann, U.: A parameterization of cirrus cloud formation: Homogeneous freezing of supercooled aerosols, *J. Geophys. Res.-Atmos.*, 107, 10, doi: 10.1029/2001jd000470, 2002.
- Kärcher, B., Dörnbrack, A., and Sölch, I.: Supersaturation Variability and Cirrus Ice Crystal Size Distributions, *J. Atmos. Sci.*, 71, 2905-2926, doi: 10.1175/JAS-D-13-0404.1, 2014.
- 965 Kärcher, B.: Formation and radiative forcing of contrail cirrus, *Nat. Commun.*, 9, 17, doi: 10.1038/s41467-018-04068-0, 2018.
- Kley, D., and Stone, E. J.: Measurement of water-vapor in the stratosphere by photo-dissociation with Ly-alpha (1216 Å) light, *Rev. Sci. Instrum.*, 49, 691-697, doi: 10.1063/1.1135596, 1978.
- Koop, T., Luo, B. P., Tsias, A., and Peter, T.: Water activity as the determinant for homogeneous ice nucleation in aqueous solutions, *Nature*, 406, 611-614, doi: 10.1038/35020537, 2000.
- 970 Krämer, M., Schiller, C., Afchine, A., Bauer, R., Gensch, I., Mangold, A., Schlicht, S., Spelten, N., Sitnikov, N., Borrmann, S., de Reus, M., and Spichtinger, P.: Ice supersaturations and cirrus cloud crystal numbers, *Atmos. Chem. Phys.*, 9, 3505-3522, 2009.
- Krämer, M., Rolf, C., Luebke, A., Afchine, A., Spelten, N., Costa, A., Meyer, J., Zöger, M., Smith, J., Herman, R. L., Buchholz, B., Ebert, V., Baumgardner, D., Borrmann, S., Klingebiel, M., and Avallone, L.: A microphysics guide to cirrus clouds – Part 1: Cirrus types, *Atmos. Chem. Phys.*, 16, 3463-3483, doi: 10.5194/acp-16-3463-2016, 2016.
- 975 Kunz, A., Schiller, C., Rohrer, F., Smit, H. G. J., Nédélec, P., and Spelten, N.: Statistical analysis of water vapour and ozone in the UT/LS observed during SPURT and MOZAIC, *Atmos. Chem. Phys.*, 8, 6603-6615, 2008.
- Kunz, A., Mueller, R., Homonnai, V., Janosi, I. M., Hurst, D., Rap, A., Forster, P. M., Rohrer, F., Spelten, N., and Riese, M.: Extending water vapor trend observations over Boulder into the tropopause region: Trend uncertainties and resulting radiative forcing, *J. Geophys. Res. Atmos.*, 118, 11269-11284, doi: 10.1002/jgrd.50831, 2013.
- 980

- Kunz, A., Spelten, N., Konopka, P., Mueller, R., Forbes, R. M., and Wernli, H.: Comparison of Fast In situ Stratospheric Hygrometer (FISH) measurements of water vapor in the upper troposphere and lower stratosphere (UTLS) with ECMWF (re)analysis data, *Atmos. Chem. Phys.*, 14, 10803-10822, doi: 10.5194/acp-14-10803-2014, 2014.
- 985 Lamquin, N., Stubenrauch, C. J., Gierens, K., Burkhardt, U., and Smit, H.: A global climatology of upper-tropospheric ice supersaturation occurrence inferred from the Atmospheric Infrared Sounder calibrated by MOZAIC, *Atmos. Chem. Phys.*, 12, 381-405, doi: 10.5194/acp-12-381-2012, 2012.
- Lee, D. S., Pitari, G., Grewe, V., Gierens, K., Penner, J. E., Petzold, A., Prather, M. J., Schumann, U., Bais, A., Bernsten, T., Iachetti, D., Lim, L. L., and Sausen, R.: Transport impacts on atmosphere and climate: Aviation, 44, 4678-4734, doi: 10.1016/j.atmosenv.2009.06.005, 2010.
- 990 Marengo, A., Thouret, V., Nédélec, P., Smit, H., Helten, M., Kley, D., Karcher, F., Simon, P., Law, K., Pyle, J., Poschmann, G., Von Wrede, R., Hume, C., and Cook, T.: Measurement of ozone and water vapor by Airbus in-service aircraft: The MOZAIC airborne program, *An overview*, *J. Geophys. Res.*, 103, 25631-25642, doi: 10.1029/98jd00977, 1998.
- May, R. D., and Webster, C. R.: Data processing and calibration for tunable diode-laser harmonic absorption spectrometers, *J. Quant. Spectrosc. Radiat. Transfer*, 49, 335-347, doi: 10.1016/0022-4073(93)90098-3, 1993.
- 995 Meyer, J., Rolf, C., Schiller, C., Rohs, S., Spelten, N., Afchine, A., Zöger, M., Sitnikov, N., Thornberry, T. D., Rollins, A. W., Bozóki, Z., Tátrai, D., Ebert, V., Kühnreich, B., Mackrodt, P., Möhler, O., Saathoff, H., Rosenlof, K. H., and Krämer, M.: Two decades of water vapor measurements with the FISH fluorescence hygrometer: a review, *Atmos. Chem. Phys.*, 15, 8521-8538, doi: 10.5194/acp-15-8521-2015, 2015.
- 1000 Müller, R., Kunz, A., Hurst, D. F., Rolf, C., Krämer, M., and Riese, M.: The need for accurate long-term measurements of water vapor in the upper troposphere and lower stratosphere with global coverage, *Earth's Future*, 4, 25-32, doi: 10.1002/2015ef000321, 2016.
- Müller, S., Hoor, P., Berkes, F., Bozem, H., Klingebiel, M., Reutter, P., Smit, H. G. J., Wendisch, M., Spichtinger, P., and Borrmann, S.: In situ detection of stratosphere-troposphere exchange of cirrus particles in the midlatitudes, *Gephys. Res. Lett.*, 42, 949-955, doi: 10.1002/2014gl062556, 2015.
- 1005 Nédélec, P., Blot, R., Boulanger, D., Athier, G., Cousin, J.-M., Gautron, B., Volz-Thomas, A., Petzold, A., and Thouret, V.: Instrumentation on commercial aircraft for monitoring the atmospheric composition on a global scale : The IAGOS system, technical overview of ozone and carbon monoxide measurements, *Tellus B*, 67, doi: 10.3402/tellusb.v67.27791, 2015.
- Neis, P., Smit, H. G. J., Krämer, M., Spelten, N., and Petzold, A.: Evaluation of the MOZAIC Capacitive Hygrometer during the airborne field study CIRRUS-III, *Atmos. Meas. Tech.*, 8, 1233-1243, doi: 10.5194/amt-8-1233-2015, 2015a.
- 1010 Neis, P., Smit, H. G. J., Rohs, S., Bundke, U., Krämer, M., Spelten, N., Ebert, V., Buchholz, B., Thomas, K., and Petzold, A.: Quality assessment of MOZAIC and IAGOS capacitive hygrometers: Insights from airborne field studies, *Tellus B*, 67, 28320, doi: 10.3402/tellusb.v67.28320, 2015b.
- Neis, P.: Water Vapour in the UTLS - Climatologies and Transport, *Forschungszentrum Jülich, Schriften des Forschungszentrums Jülich, Reihe Energie und Umwelt FZJ-2017-07862*, 124 pp., 2017.
- 1015 Pan, L. L., Bowman, K. P., Atlas, E. L., Wofsy, S. C., Zhang, F. Q., Bresch, J. F., Ridley, B. A., Pittman, J. V., Homeyer, C. R., Romashkin, P., and Cooper, W. A.: The Stratosphere-Troposphere Analyses of Regional Transport 2008 experiment, *Bull. Amer. Meteorol. Soc.*, 91, 327-342, doi: 10.1175/2009bams2865.1, 2010.
- Penner, J. E., Zhou, C., Garnier, A., and Mitchell, D. L.: Anthropogenic Aerosol Indirect Effects in Cirrus Clouds, *J. Geophys. Res.-Atmos.*, 123, 11652-11677, doi: 10.1029/2018jd029204, 2018.
- 1020 Petetin, H., Jeoffrion, M., Sauvage, B., Athier, G., Blot, R., Boulanger, D., Clark, H., Cousin, J.-M., Gheusi, F., Nedelec, P., Steinbacher, M., and Thouret, V.: Representativeness of the IAGOS airborne measurements in the lower troposphere, *Elem Sci Anth.*, 6, 23, doi: 10.1525/elementa.280, 2018.
- Petzold, A., Thouret, V., Gerbig, C., Zahn, A., Brenninkmeijer, C. A. M., Gallagher, M., Hermann, M., Pontaud, M., Ziereis, H., Boulanger, D., Marshall, J., Nédélec, P., Smit, H. G. J., Frieß, U., Flaud, J.-M., Wahner, A., Cammas, J.-P., Volz-Thomas, A., and IAGOS-Team: Global-Scale Atmosphere Monitoring by In-Service Aircraft – Current Achievements and Future Prospects of the European Research Infrastructure IAGOS, *Tellus B*, 67, 28452, doi: 10.3402/tellusb.v67.28452, 2015.
- 1025 Petzold, A., Krämer, M., Neis, P., Rolf, C., Rohs, S., Berkes, F., Smit, H. G. J., Gallagher, M., Beswick, K., Lloyd, G., Baumgardner, D., Spichtinger, P., Nedelec, P., Ebert, V., Buchholz, B., Riese, M., and Wahner, A.: Upper tropospheric water vapour and its interaction with cirrus clouds as seen from IAGOS long-term routine in situ observations, *Faraday Discuss.*, 200, 229-249, doi: 10.1039/c7fd00006e, 2017.
- Pruppacher, H. R., and Klett, J. D.: *Microphysics of Clouds and Precipitation*, 2nd ed., Kluwer Academic Publishers, AA Dordrecht, 1997.
- 1035 Reichler, T., Dameris, M., and Sausen, R.: Determining the tropopause height from gridded data, *Geophys. Res. Lett.*, 30, doi: 10.1029/2003gl018240, 2003.
- Reutter, P., Neis, P., Rohs, S., and Sauvage, B.: Ice supersaturated regions: properties and validation of ERA-Interim reanalysis with IAGOS in situ water vapour measurements, *Atmos. Chem. Phys.*, 20, 787-804, doi: 10.5194/acp-20-787-2020, 2020.
- 1040 Riese, M., Ploeger, F., Rap, A., Vogel, B., Konopka, P., Dameris, M., and Forster, P.: Impact of uncertainties in atmospheric mixing on simulated UTLS composition and related radiative effects, *J. Geophys. Res.-Atmos.*, 117, D16305, doi: 10.1029/2012jd017751, 2012.
- Rolf, C., Vogel, B., Hoor, P., Afchine, A., Günther, G., Krämer, M., Müller, R., Müller, S., Spelten, N., and Riese, M.: Water vapor increase in the lower stratosphere of the Northern Hemisphere due to the Asian monsoon anticyclone observed during the TACTS/ESMVal campaigns, *Atmos. Chem. Phys.*, 18, 2973-2983, doi: 10.5194/acp-18-2973-2018, 2018.
- 1045 Santee, M. L., Manney, G. L., Livesey, N. J., Schwartz, M. J., Neu, J. L., and Read, W. G.: A comprehensive overview of the climatological composition of the Asian summermonsoon anticyclone based on 10 years of Aura Microwave Limb Sounder measurements, *J. Geophys. Res.-Atmos.*, 122, 5491-5514, doi: 10.1002/2016jd026408, 2017.

- 1050 Schwartz, M. J., Read, W. G., Santee, M. L., Livesey, N. J., Froidevaux, L., Lambert, A., and Manney, G. L.: Convectively injected water vapor in the North American summer lowermost stratosphere, *Geophys. Res. Lett.*, 40, 2316-2321, doi: 10.1002/grl.50421, 2013.
- Seidel, D. J., Berger, F. H., Diamond, H. J., Dykema, J., Goodrich, D., Immler, F., Murray, W., Peterson, T., Sisterson, D., Sommer, M., Thorne, P., Voemel, H., and Wang, J.: Reference Upper-Air Observations for Climate: Rationale, Progress, and Plans, *Bull. Am. Met. Soc.*, 90, 361-+, doi: 10.1175/2008bams2540.1, 2009.
- 1055 Sitnikov, N. M., Yushkov, V. A., Afchine, A. A., Korshunov, L. I., Astakhov, V. I., Ulanovskii, A. E., Krämer, M., Mangold, A., Schiller, C., and Ravegnani, F.: The FLASH instrument for water vapor measurements on board the high-altitude airplane, *Instrum. Exp. Tech.*, 50, 113-121, doi: 10.1134/s0020441207010174, 2007.
- Smit, H. G. J., Volz-Thomas, A., Helten, M., Paetz, W., and Kley, D.: An in-flight calibration method for near-real-time humidity measurements with the airborne MOZAIC sensor, *J. Atmos. Oceanic Technol.*, 25, 656-666, doi: 10.1175/2007jtecha975.1, 2008.
- 1060 Smit, H. G. J., Rohs, S., Neis, P., Boulanger, D., Krämer, M., Wahner, A., and Petzold, A.: Technical Note: Reanalysis of upper troposphere humidity data from the MOZAIC programme for the period 1994 to 2009, *Atmos. Chem. Phys.*, 14, 13241-13255, doi: 10.5194/acp-14-13241-2014, 2014.
- Sonntag, D.: Advances in the field of hygrometry, *Meteorol. Z.*, N.F. 3, 51-66, 1994.
- 1065 Spang, R., Gunther, G., Riese, M., Hoffmann, L., Muller, R., and Griessbach, S.: Satellite observations of cirrus clouds in the Northern Hemisphere lowermost stratosphere, *Atmos. Chem. Phys.*, 15, 927-950, doi: 10.5194/acp-15-927-2015, 2015.
- Spichtinger, P., Gierens, K., and Read, W.: The statistical distribution law of relative humidity in the global tropopause region, *Meteorol. Z.*, 11, 83-88, doi: 10.1127/0941-2948/2002/0011-0083, 2002.
- Spichtinger, P., Gierens, K., Leiterer, U., and Dier, H.: Ice supersaturation in the tropopause region over Lindenberg, Germany, *Meteorol. Z.*, 12, 143-156, doi: 10.1127/0941-2948/2003/0012-0143, 2003a.
- 1070 Spichtinger, P., Gierens, K., and Read, W.: The global distribution of ice-supersaturated regions as seen by the Microwave Limb Sounder, *Q. J. R. Meteorol. Soc.*, 129, 3391-3410, doi: 10.1256/qj.02.141, 2003b.
- Spichtinger, P., Gierens, K., and Wernli, H.: A case study on the formation and evolution of ice supersaturation in the vicinity of a warm conveyor belt's outflow region, *Atmos. Chem. Phys.*, 5, 973-987, 2005.
- 1075 Spichtinger, P., and Leschner, M.: Horizontal scales of ice-supersaturated regions, 68, doi: 10.3402/tellusb.v68.29020, 2016.
- Stubenrauch, C. J., Cros, S., Guignard, A., and Lamquin, N.: A 6-year global cloud climatology from the Atmospheric InfraRed Sounder AIRS and a statistical analysis in synergy with CALIPSO and CloudSat, *Atmos. Chem. Phys.*, 10, 7197-7214, doi: 10.5194/acp-10-7197-2010, 2010.
- Stubenrauch, C. J., Rossow, W. B., Kinne, S., Ackerman, S., Cesana, G., Chepfer, H., Di Girolamo, L., Getzewich, B., Guignard, A., Heidinger, A., Maddux, B. C., Menzel, W. P., Minnis, P., Pearl, C., Platnick, S., Poulsen, C., Riedi, J., Sun-Mack, S., Walther, A., Winker, D., Zeng, S., and Zhao, G.: Assessment of Global Cloud Datasets from Satellites: Project and Database Initiated by the GEWEX Radiation Panel, *Bull. Amer. Meteorol. Soc.*, 94, 1031-1049, doi: 10.1175/bams-d-12-00117.1, 2013.
- 1080 Stuber, N., Forster, P., Radel, G., and Shine, K.: The importance of the diurnal and annual cycle of air traffic for contrail radiative forcing, *Nature*, 441, 864-867, doi: 10.1038/nature04877, 2006.
- 1085 Thouret, V., Cammas, J. P., Sauvage, B., Athier, G., Zbinden, R., Nédélec, P., Simon, P., and Karcher, F.: Tropopause referenced ozone climatology and inter-annual variability (1994-2003) from the MOZAIC programme, *Atmos. Chem. Phys.*, 6, 1033-1051, doi: 10.5194/acp-6-1033-2006, 2006.
- WMO: Meteorology - a three-dimensional science, *WMO Bull.*, 6, 134-138, 1957.
- 1090 Zahn, A., and Brenninkmeijer, C. A. M.: New directions: A chemical tropopause defined, *Atmos. Environ.*, 37, 439-440, doi: 10.1016/s1352-2310(02)00901-9, 2003.
- Zahn, A., Christner, E., van Velthoven, P. F. J., Rauthe-Schoech, A., and Brenninkmeijer, C. A. M.: Processes controlling water vapor in the upper troposphere/lowermost stratosphere: An analysis of 8 years of monthly measurements by the IAGOS-CARIBIC observatory, *J. Geophys. Res. Atmos.*, 119, 11505-11525, doi: 10.1002/2014jd021687, 2014.
- 1095 Zöger, M., Afchine, A., Eicke, N., Gerhards, M. T., Klein, E., McKenna, D. S., Morschel, U., Schmidt, U., Tan, V., Tuitjer, F., Woyke, T., and Schiller, C.: Fast in situ stratospheric hygrometers: A new family of balloon-borne and airborne Lyman alpha photofragment fluorescence hygrometers, *J. Geophys. Res.*, 104, 1807-1816, doi: 10.1029/1998jd100025, 1999.