

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 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 H_2O 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 H_2O VMR up to the tropopause layer. Across the tropopause layer, H_2O 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 H_2O 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 - O₃ 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 O₃ 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, H₂O VMR and O₃ 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 O₃ 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).

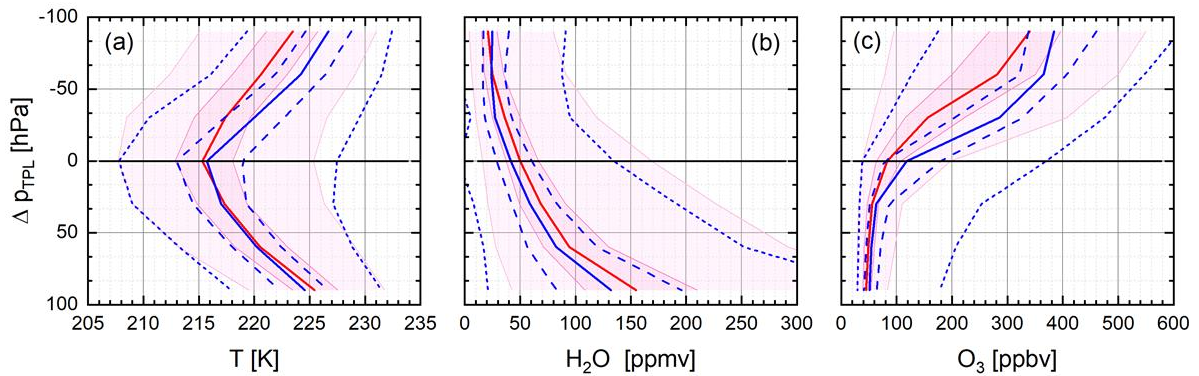


Figure 9. Vertical distribution of temperature T (a), H₂O VMR (b), and ozone VMR (c) relative to the 2 PVU dynamical tropopause and to the thermal tropopause; vertical distributions relative to the thermal tropopause are presented as percentiles [1, 25, 50, 75, and 99] by blue lines and relative to the 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 #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- σ 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- σ 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, O₃ VMR, H₂O VMR, RH_{ice}, 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_{ice} >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_{ice} >103%, or even RH_{ice} >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_{ice} data with a reported offset of $-0.15 \pm 1.1.29 \%$ RH_L (Neis et al. 2015a). From these results we conclude that we don't see a systematic bias in the RH_{ice} data reported from the MCH instrument. Hence, we assume a random scatter of the RH_{ice} data around the zero value of RH_{ice} = 100% which justifies the assumption of RH_{ice} = 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, H₂O VMR and O₃ VMR over the North Atlantic. The new Figure 9 presents the results:

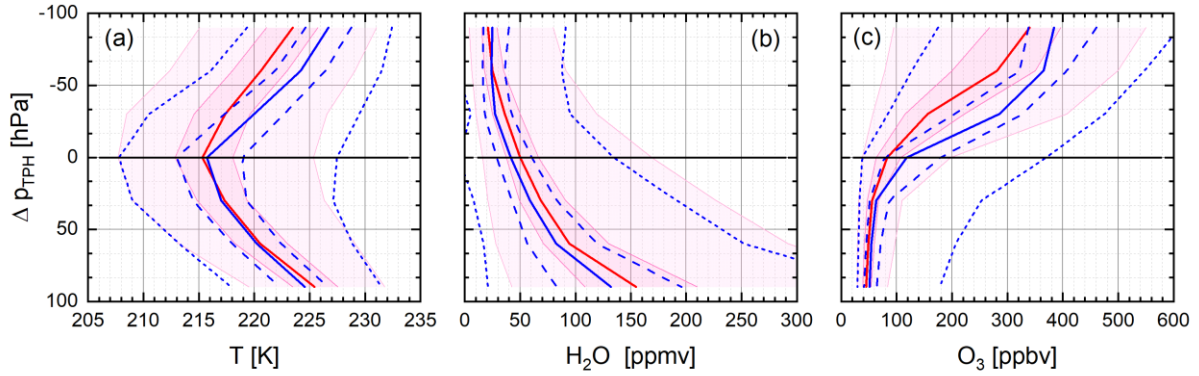


Figure 9. Vertical distribution of temperature T (a), H₂O VMR (b), and O₃ VMR (c) relative to the 2 PVU dynamical tropopause and to the thermal tropopause; vertical distributions relative to the thermal tropopause are presented as percentiles [1, 25, 50, 75, and 99] by blue lines and relative to the 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 O₃ 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 O₃ 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 H₂O VMR values have been obtained. We introduced on sentence in Section 2.1: “The H₂O VMR was finally calculated from the simultaneously measured RH_{ice} 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 O₃, RH_{ice} and temperature, and H₂O VMR deduced from RH_{ice} 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- α 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- σ criterion (95% confidence level), we obtain a MCH limit of detection (LOD) of RH_{ice,LOD} = 10% which again transfers into a minimum detectable H₂O 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_{ice} uncertainty so

that the precision of H₂O 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 H₂O VMR deeper in the stratosphere and implies a limited use of the MOZAIC H₂O sensor in the stratosphere dominated by low RH_{ice} 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_{ice}. 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_{ice}. 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 H₂O VMR and temperatures above the thermal tropopause in the summer months, whereas RH_{ice} 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_{ice} with the IFC applied, so please make this clear.*

Authors: The figure caption reads now: “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).”

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, H₂O VMW and RH_{ice} is shown in Fig. 8.

The source of H₂O VMR measurements is introduced in: "The H₂O VMR was finally calculated from the simultaneously measured RH_{ice} 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 $TP_{press} - 15$ hPa and $TP_{press} - 45$ hPa? 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 $p_{therm.TPH} = 0$ hPa for the tropopause layer (TPL) itself and then at $p_{therm.TPH} \pm 30$ hPa, $p_{therm.TPH} \pm 60$ hPa, and finally at $p_{therm.TPH} \pm 90$ hPa. From this vertical spacing, the separation of air masses is achieved by applying the following criteria (formulated for the thermal tropopause only):

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

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

UT : $p > p_{therm.TPH} + 15$ hPa; limited to lower altitudes by $p < 350$ 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, H₂O VMR further decreases in case of non-ISSR conditions. For ISSR conditions, however, H₂O VMR remains constant with height throughout the layer just above the tropopause. Doubling of H₂O 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 H₂O 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 H₂O 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 O₃ 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_{ice} 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_{ice} – RH_{ice} 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 Δ 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)."

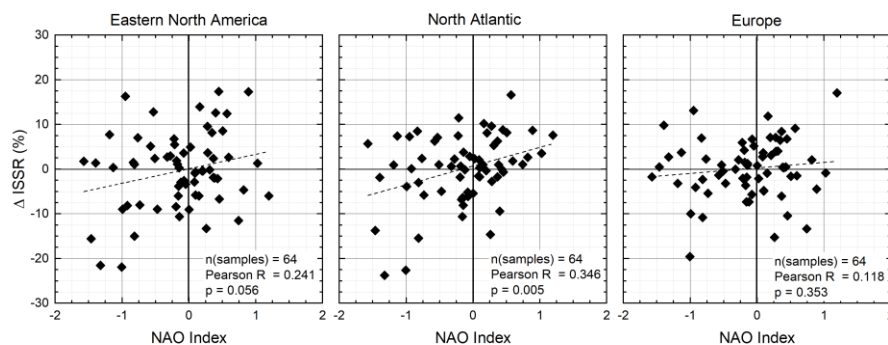


Figure 16. Correlation analysis with respect to the correlation of signs between NAO index and deviation of ISSR occurrence from the long-term average (Δ ISSR) for the target regions; numbers indicate the results from the correlation analysis with respect to number of samples n, Pearson R and significance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

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

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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 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 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 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 H_2O 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 H₂O volume-mixing-ratio (VMR) up to the thermal-tropopause layer. Across the tropopause layer, H₂O 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 H₂O 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_{ice} 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_{ice} > 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 H₂O 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-CO₂ 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_{ice} 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_{ice} 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_{ice} 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 H₂O, O₃, 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 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_{ice} (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⁻¹.

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.

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⁻¹ 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 = 10⁻⁶ m² K s⁻¹ kg⁻¹. 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~~

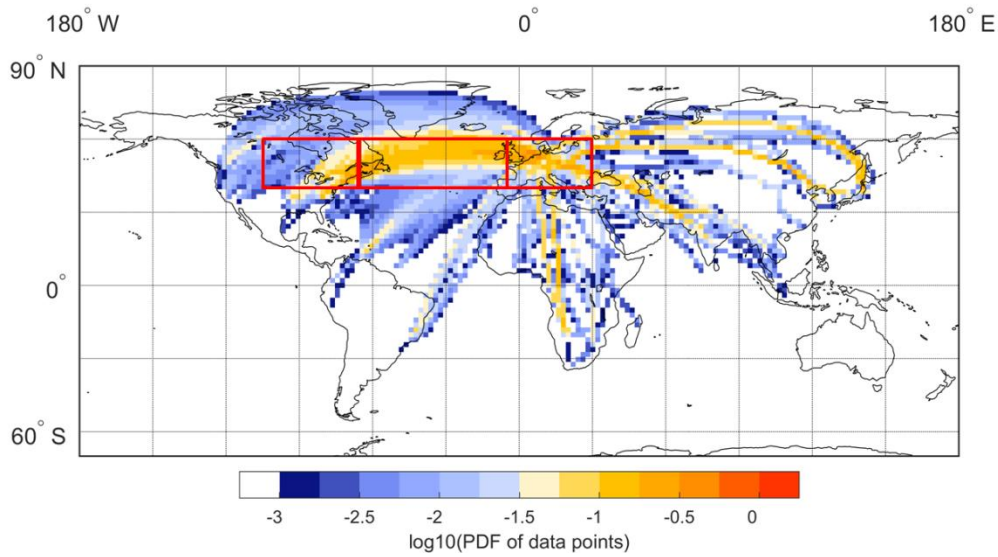


Figure 1. Global coverage of water vapour observations by MOZAIC for the period 1995 to 2010, shown as decadal logarithm of the probability distribution function (PDF) of the data points (fraction of measurements in a certain grid box); red boxes indicate the target areas for our analyses.

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 \(temperature, O₃ VMR, H₂O VMR, RH_{ice}, ISSR fraction\) 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 O₃ 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 α 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_{ice} 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 H₂O VMR was finally calculated from the simultaneously measured RH_{ice} 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- σ criterion (95% confidence level), the MCH limit of detection (LOD) is RH_{ice,LOD} = 12% which transfers into a minimum detectable H₂O 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- α 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- σ 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 ± 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

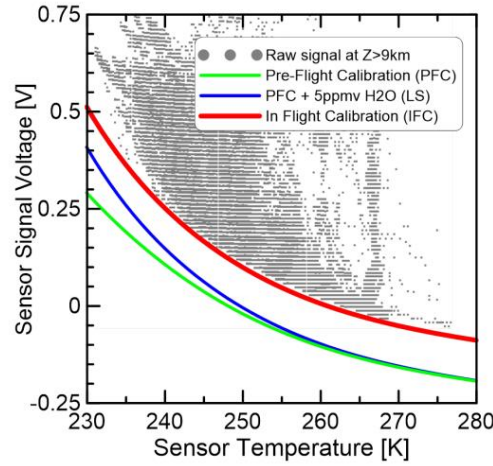


Figure 2. Raw signal of the MOZAIC humidity sensor aboard one MOZAIC aircraft as a function of the sensor temperature inside the aeronautic housing obtained at cruise altitude ($z = 9 - 12$ km). Green line: zero signal from pre-flight calibration (PFC); blue line: superposition of zero signal from PFC 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.~~

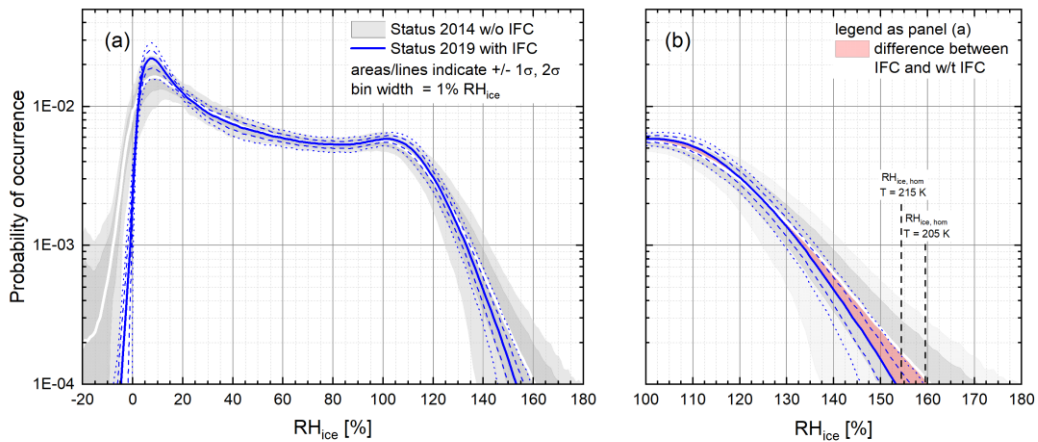


Figure 3. Averaged probability density functions of RH_{ice} for the entire MOZAIC period from 1995 to 2010 (a), and the zoom into the region of ice-supersaturation (b); data stem from the reanalysis (Smit et al., 2014) without (white line, grey areas), and with (blue lines) the in-flight calibration 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 2σ - 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.

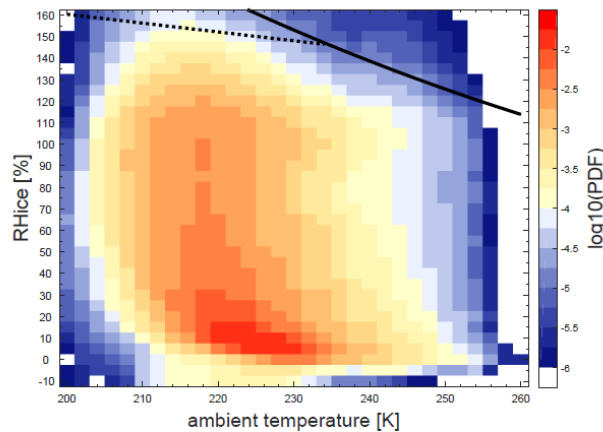


Figure 4. Distribution of RH_{ice} for the entire MOZAIC period from 1995 to 2010 with IFC applied as a function of ambient temperature with the colour indicating the probability of occurrence; the lines represent water saturation (solid line; Sonntag, 1994) and the threshold RH_{ice} for homogeneous 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- α 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.

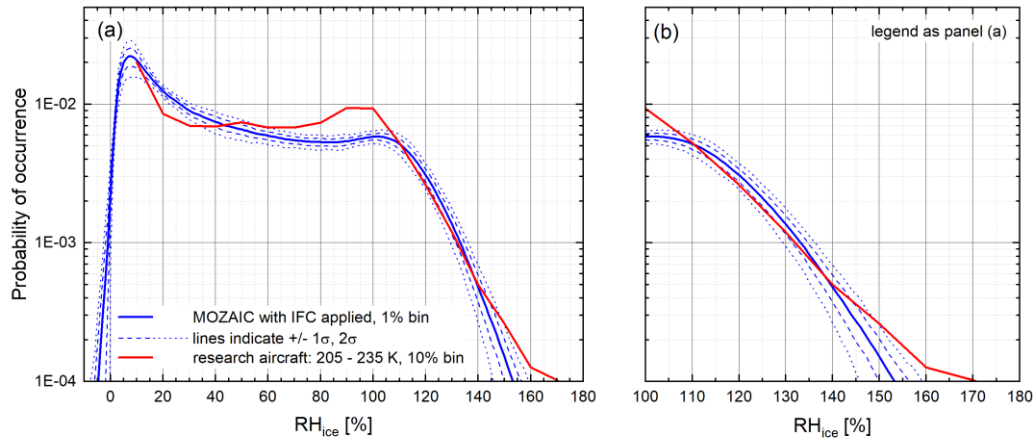


Figure 5. Averaged probability density functions of RH_{ice} for the entire MOZAIC period from 1995 to 2010; with the in-flight calibration method applied (blue lines) and respective RH_{ice} PDF from 250 research aircraft flights collected in the Juelich In-situ Airborne Database (Krämer et al., 2016).

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 H₂O 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 H₂O VMR values in winter and spring months and a maximum H₂O 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.

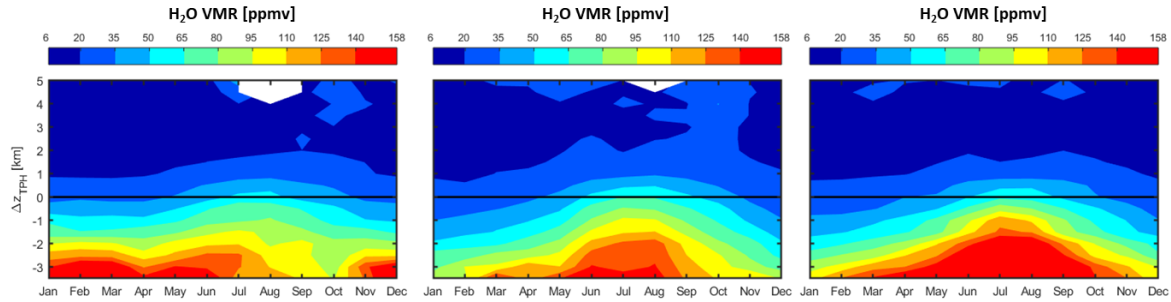


Figure 6. 15-year averaged annual cycles of H₂O VMR vertical distributions of H₂O VMR for latitudes 40 °N to 60 °N and for the regions (from left to right) Eastern North America (105 °W to 65 °W), North Atlantic (65 °W to 5 °W) and Europe (5 °W to 30 °E).

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 H₂O 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 H₂O VMR was reported by Zahn et al. (2014) from zonal-averaged H₂O VMR observations by the CARIBIC system. In contrast to MOZAIC, the CARIBIC H₂O 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 H₂O 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 H₂O 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).

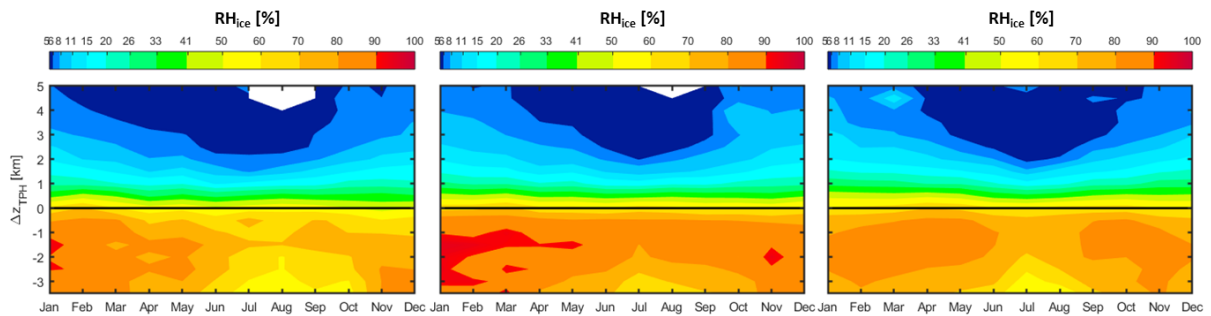


Figure 7. 15-year averaged annual cycles of RH_{ice} for latitudes 40 °N to 60 °N and for the regions (from left to right) Eastern North America (105 °W to 65 °W), North Atlantic (65 °W to 5 °W) and Europe (5 °W to 30 °E).

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.

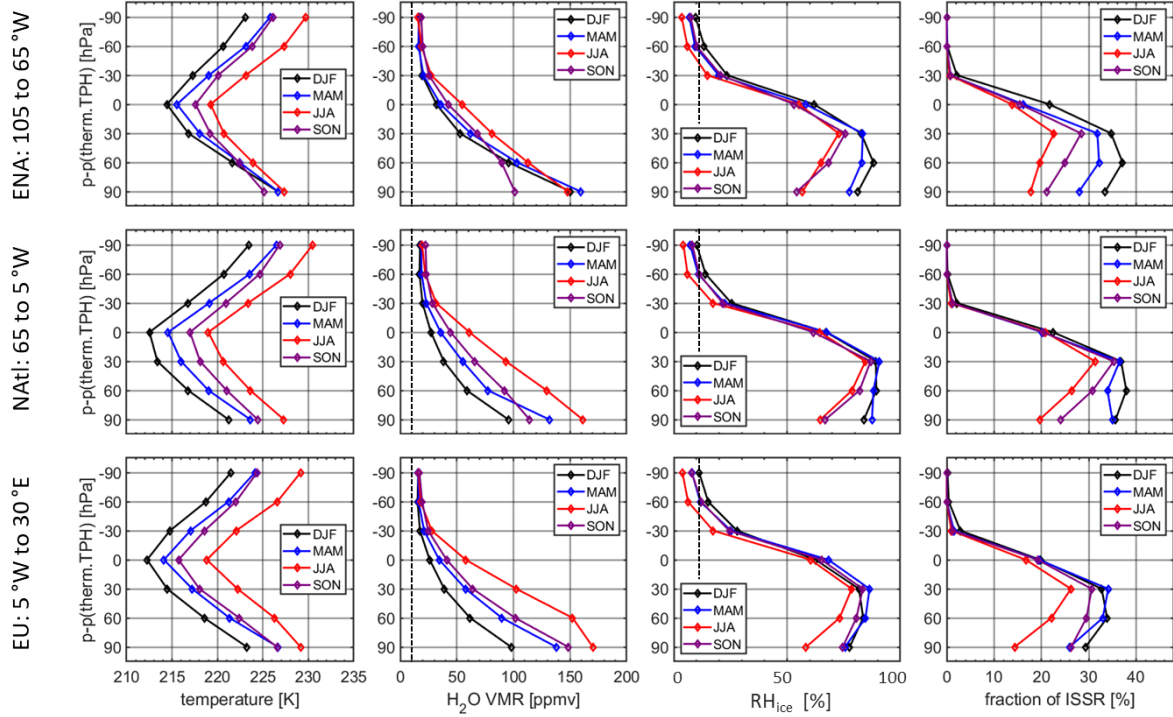


Figure 8. Vertical distribution of mean temperature, H_2O mixing ratio, RH_{ice} and fraction of ice-supersaturated regions (ISSR) for seven pressure layers around the thermal tropopause; layer thickness is 30 hPa and layers are spaced equally relative to the tropopause pressure level; dotted lines indicate the MCH 2- σ 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 $\Delta p = p_{layer} - p_{therm.TPH}$.

Δp (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 $\Delta p = p_{layer} - p_{therm.TPH}$.

Δp	DJF	MAM	JJA	SON	AVG(ENA, Natl, EU)
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(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 - O₃ 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 O₃ 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, H₂O VMR and O₃ 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 O₃ 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).

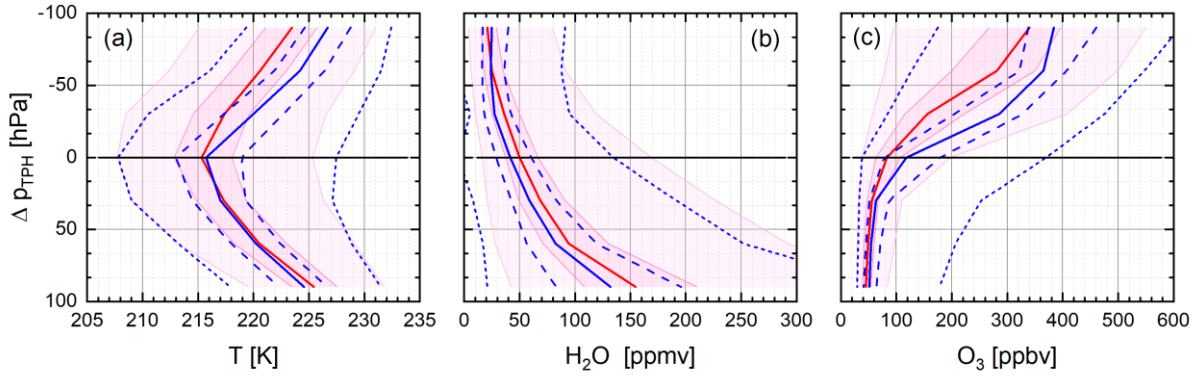


Figure 9. Vertical distribution of temperature T (a), H_2O VMR (b), and O_3 VMR (c) relative to the 2 PVU dynamical tropopause and to the thermal tropopause; vertical distributions relative to the thermal tropopause are presented as percentiles [1, 25, 50, 75, and 99] by blue lines and relative to the 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 [%]
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		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 H₂O 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 sub-saturated 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 sub-saturated 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 H₂O VMR for ice-supersaturated and ice-sub-saturated regions with exponentially decreasing absolute humidity up to the tropopause layer. Above both tropopause layers, H₂O VMR ~~is~~ further decreases ~~for their case of~~ non-ISSR conditions. For ISSR conditions, however, H₂O 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 H₂O 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 H₂O 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.

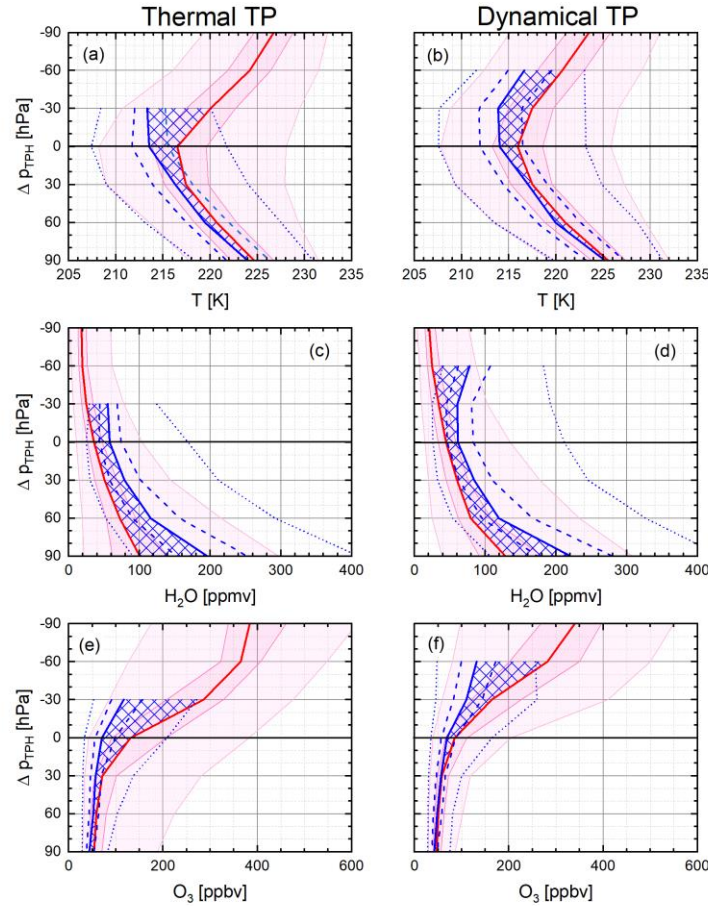


Figure 10. Vertical distribution of temperature, H_2O VMR, and ozone VMR for ISSR relative to the thermal tropopause height (panels a, c and e) and 2 PVU dynamical tropopause height (panels b, d, and f). ISSR conditions are presented as percentiles [1, 25, 50, 75, and 99] by blue lines and non-ISSR conditions 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 ~~ir~~ 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.

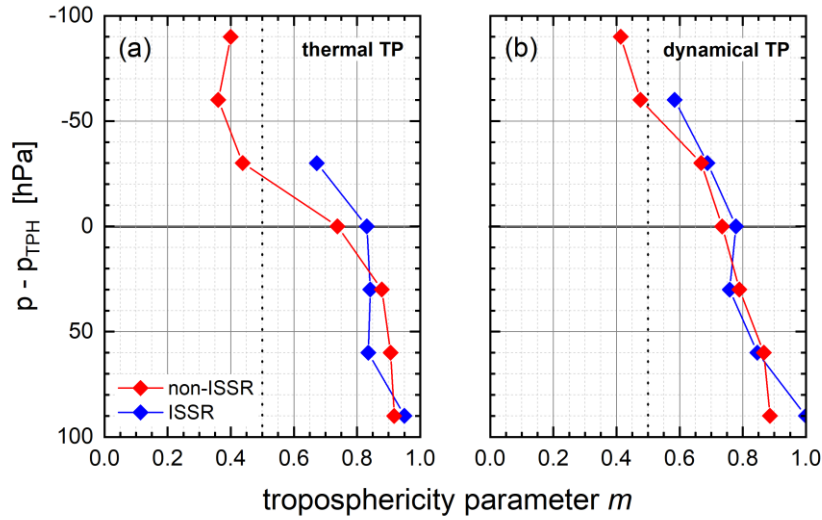


Figure 11. Vertical distribution of the troposphericity parameter m for ISSR and non-ISSR air masses with respect to the thermal (a) and dynamical (b) tropopause.

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).

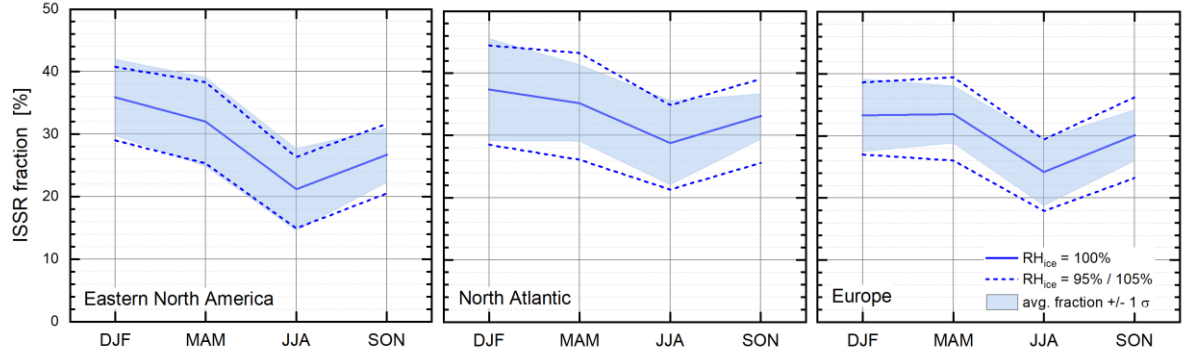


Figure 12. Annual cycles of ISSR occurrence shown as occurrence probability for $RH_{ice} > 100\%$, for the regions Eastern North America, North Atlantic and Europe; considered years are 1995 to 2010, with shaded areas representing probabilities for the average value (thick lines) $\pm 1\sigma$, and 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.

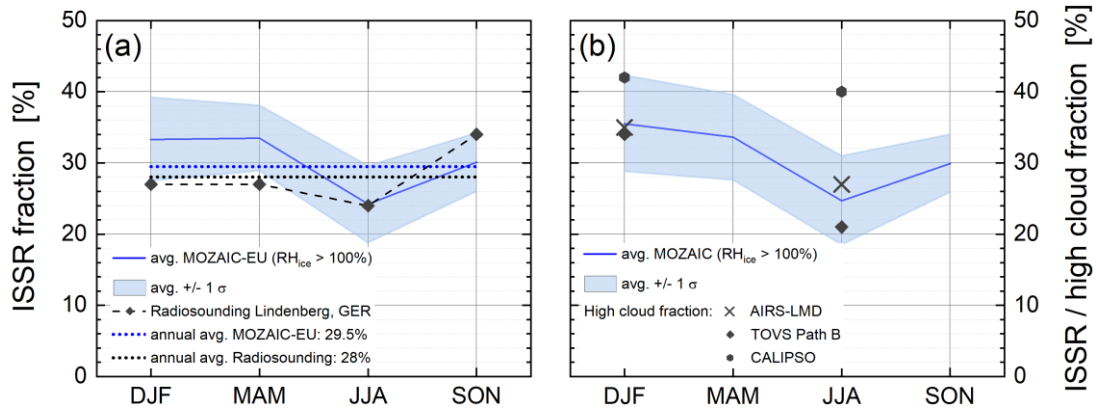


Figure 13. a: Seasonal cycle of ISSR occurrence probability, i.e. $p(RH_{ice} > 100\%)$, averaged over Europe for the years 1995 to 2010 for the two UT layers positioned closest to the thermal tropopause; symbols represent the annual cycle of the Lindenberg sounding (2000 – 2001) from Spichtinger et 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 this

~~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.~~

690 The ~~close match~~good agreement between MOZAIC in-situ observations of ~~RH_{ice} -ISSR occurrence~~ with the high-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-
 715 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).

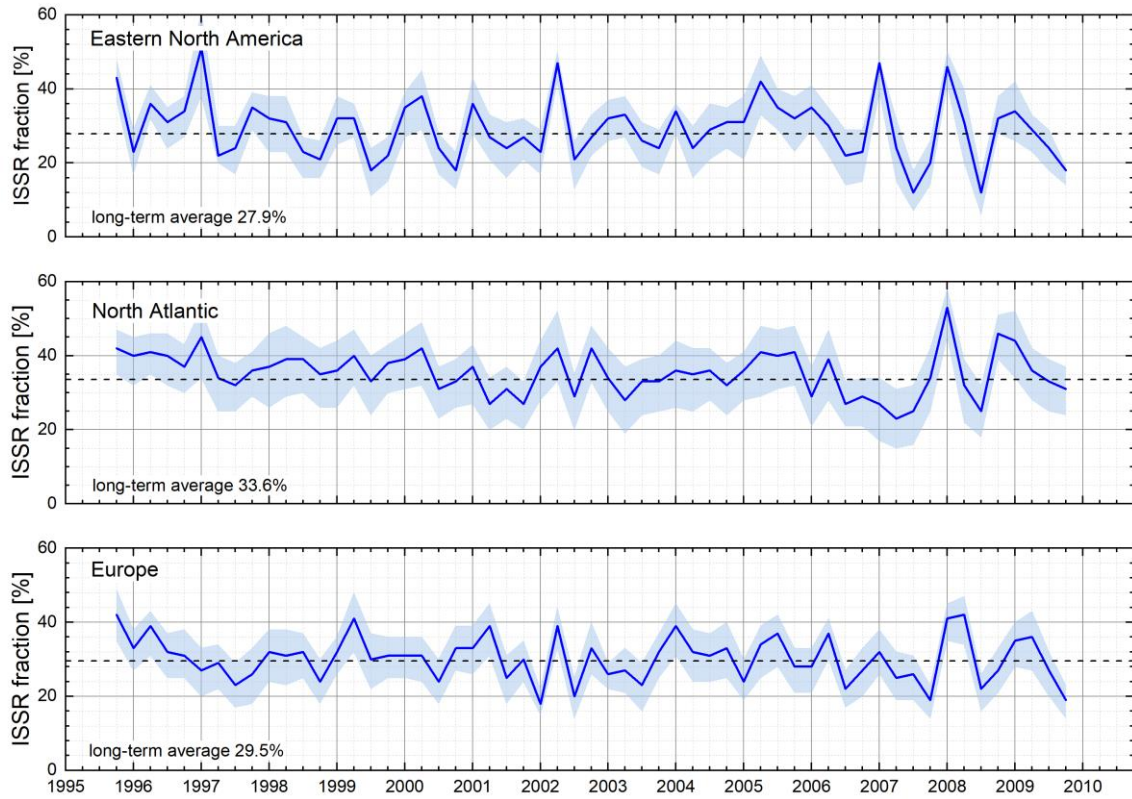


Figure 14. Time series of ISSR fraction (probability of occurrence) for latitudes 40°N to 60°N and for the
 715 regions (from top to bottom) Eastern North America (105°W to 65°W), North Atlantic (65°W to 5°W) and
 Europe (5°W to 30°E) for the top UT layer, situated just below the tropopause layer; with 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.

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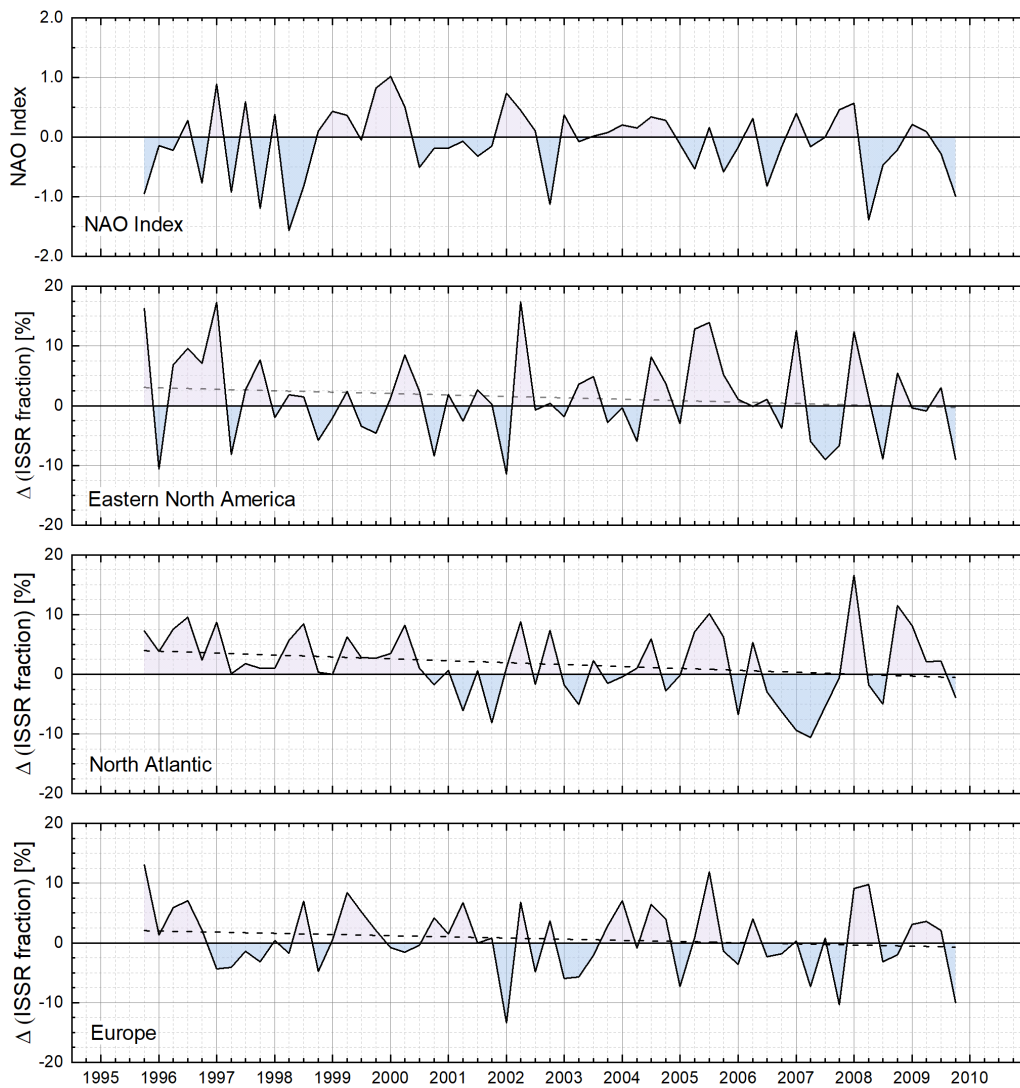


Figure 15. De-seasonalised time series of ISSR fraction (probability of occurrence) for latitudes 40°N to 60°N and for the regions (from top to bottom) Eastern North America (105°W to 65°W), North Atlantic (65°W to 5°W) and Europe (5°W to 30°E).

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 Δp (Iceland L to Azores H)

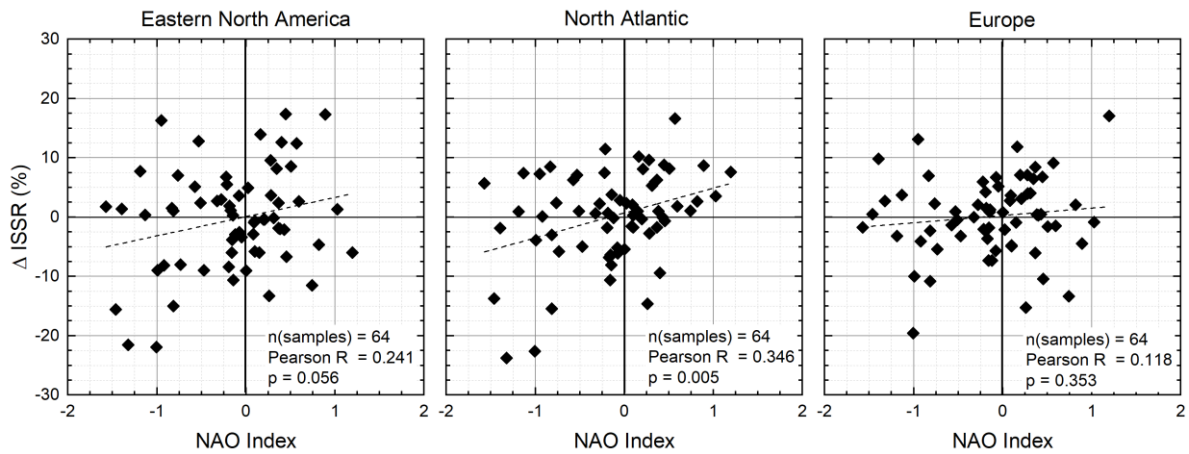


Figure 16. Cluster-Correlation analysis with respect to the correlation of signs between NAO index and deviation of ISSR occurrence from the long-term average (Δ ISSR) for the target regions; numbers indicate the results from the correlation analysis with respect to number of samples n, Pearson R and significance level p; black symbols and grey shaded clusters indicate the same sign for NAO index and Δ 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 (Δ 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 Δ 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 Δ 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 Δ 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 Δ 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_{ice} 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_{ice} 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 H₂O_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.

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