

General Remarks

We thank both reviewers for their time and the constructive comments, which improve the quality of our manuscript significantly. The referee comments are formatted in grey and our reply in black with indentation and numbering (R##). The line and figure numbers refer to the revised version.

Specific Reply to Referee #1

This manuscript presents the role of aerosol and dynamics on ice formation in stratiform mixed-phase clouds based on the comparison of ground-based remote-sensing stations (one in the South hemisphere and two in the North hemisphere). The topic is well introduced. Despite the methodology is complex, the authors have made a good work, being quite easy to follow. Results and discussion sections are also well structured, helping to identify the main findings. Additionally, the paper is written in good English and, from my point of view, this paper makes an excellent contribution to the research field of aerosol-cloud interaction. Therefore, I suggest the acceptance of this manuscript with minor revisions.

Minor comments:

The title seems to attribute the contrasts in ice formation to the hemispheric location of the stations. Punta Arenas (South) vs Limassol and Leipzig (North). However, I would say that the contrasts can be finally attributed to 1) pristine vs 'dirty' atmospheric conditions and the gravity wave occurrences. I mean, a similar station (as Punta Arenas) but located in the North hemisphere would not show similar results?

R1: That's an interesting point, raised by Referee #1. Indeed, one of the next steps would be to locate a northern-hemispheric mid-latitude site, which shows similar dynamic conditions as Punta Arenas while being exposed to the high northern-hemispheric aerosol concentrations. Perhaps the lee of the Scandinavian mountains (Scotland, Sweden, Finland) would be an option. Or some place in north-western parts of North America. Similarly, we are indeed already preparing for a remote-sensing campaign in southern New Zealand, where we expect stronger perturbations of the aerosol load caused by emissions of the Australian continent, while orographic effects are expected to be similar.

P4 L114-L117: The phrase 'Afterwards, the methods for cloud selection and vertical velocity characterization are introduced.' Seems to be redundant.

R2: We corrected the text at the respective position.

P4 L120 (suggestion): Boundary layer coupling and vertical dynamics are discussed in Sec. 3.2.2 and Sec. 3.2.3, respectively.

R3: Included as suggested.

P7 L169: Cloud Top Temperature is crucial in this paper. However, it is obtained from models (ECMWF's IFS analysis and GDAS) instead of from the microwave radiometer. Why? Which is the uncertainty of the Cloud Top Temperature? Since findings (chapter 5) are formulated at -5, -10, -15, -20°C, the uncertainty should be below 5°C

R4: Similar to our previous studies (see, e.g., Seifert et al., 2010, Sec. 2.3; Kanitz et al., 2011, Sec. 2), we checked for the accuracy of the model temperatures by comparing the model values to available co-located radiosondes. In agreement to what was found previously, the uncertainties in the free troposphere (2-10 km) for Punta Arenas and Limassol are in the order of 1 K. Higher deviations can be expected in the boundary layer and in the tropopause

region. Nevertheless, as also suggested by Referee #2, we checked for the impact of the temperature uncertainty on the frequency of ice-formation statistics with a Monte Carlo sampling approach. A random error drawn from a uniform distribution between -2 and 2 K is added to the CTT. The resulting statistics are shown in Fig. 1 below. The findings described in the manuscript also hold for a 4 K uncertainty in the temperature.

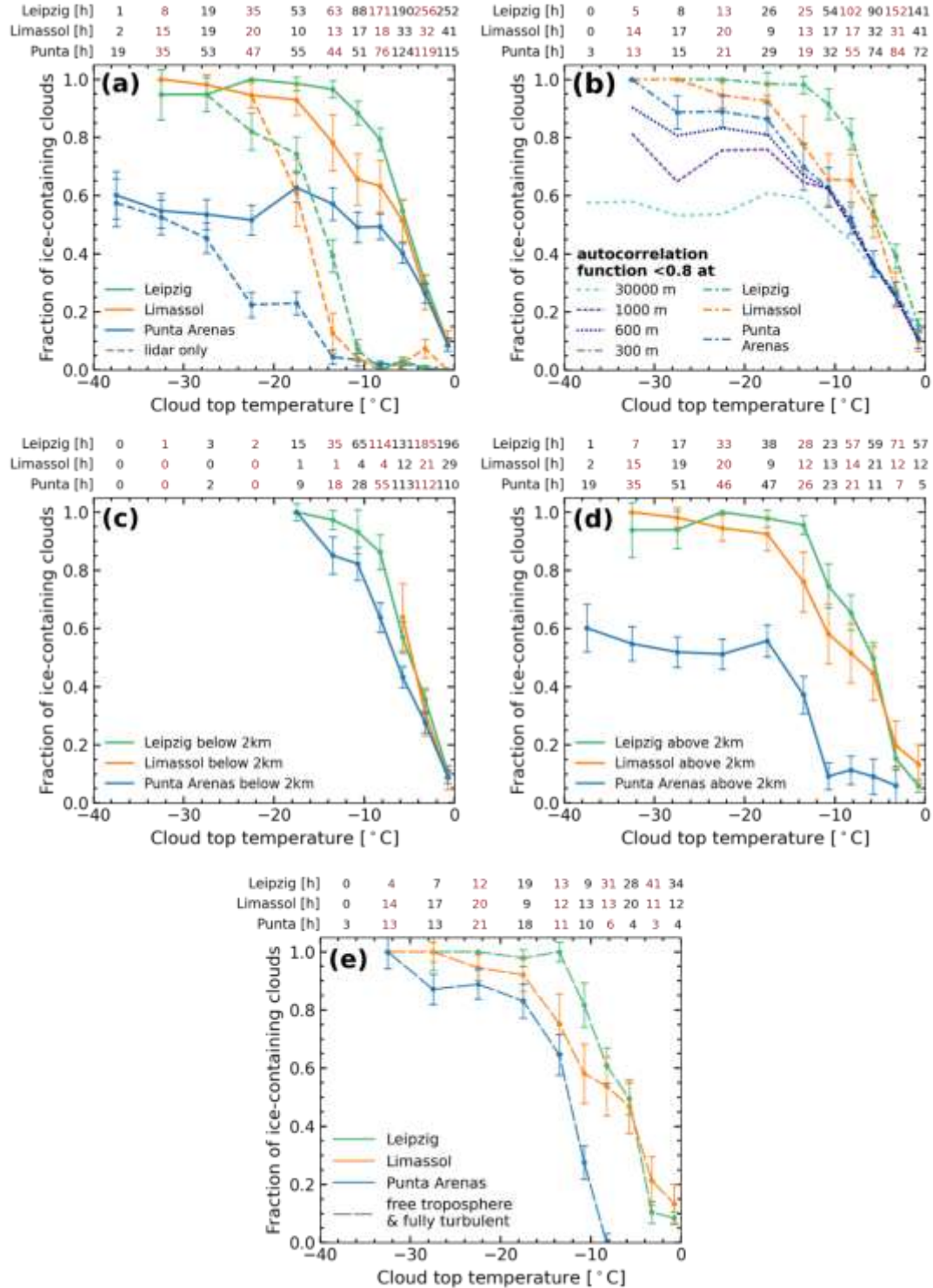


Fig 1: Impact of random errors in CTT on fraction of ice containing clouds at Leipzig, Limassol, and Punta Arenas similarly to the figures shown in the manuscript. (a) basic statistics and lidar-extinction threshold, (b) filtered for wave clouds with the autocorrelation threshold, (c) cloud base below 2 km, (d) cloud base above 2 km, and (e) for free tropospheric and fully turbulent clouds.

Specific Reply to Referee #2

Summary:

Radenz et al. use a comprehensive suite of observations for three contrasting ground-based locations in order to quantify and understand differences in ice formation. They conduct a comprehensive analysis of aerosol and cloud properties derived from the ground-based sensors in order to reach four main conclusions: (1) average backscatter is fairly close between sites; (2) ice formation at relatively warm temperatures is more common than previously thought; (3) clouds coupled to the aerosol-rich BL show more ice formation; (4) gravity waves form liquid clouds over Punta Arenas and bias the cold statistics. I commend the authors for their thorough analysis and note that the four key points I summarised above (From their abstract and conclusions) are all important conclusions.

Although I suggest 'major revision' due to one major comment directly below, I think that this can easily be implemented (along with the minor comments).

I would strongly support acceptance of the revised manuscript by ACP. It will be a very good contribution to the literature.

Major comment:

Line 263. On the use of the Fast Fourier Transform (FFT). The authors described how the vertical velocity time-series is the input into the FFT in order to calculate power spectral density. However, in lines 253-254, the authors state that 'vertical velocity is sampled at the pixel with the maximum backscatter out of the heights...', and illustrate this as the dashed line in Figure 2c. Note that this varies in height. In other words, the authors are mixing a height and time series as use for input for the FFT. The series for input into the FFT should be time- only: it is not correct to incorporate information from a range of altitudes in your time-series. While I suspect these effects are likely small and won't change the conclusions, the authors should reprocess their data, using the vertical velocity at a constant altitude, for each cloud event. I leave it to Radenz et al. to choose what the altitude is for each cloud, but if you would like a suggestion, then perhaps use the mean (or median) altitude of peak backscatter for each cloud. Please propagate these changes through the manuscript.

R5: Thanks for the suggestion. We originally implemented the height-adaptive sampling at the maximum of the backscatter signal to avoid gaps in the timeseries. We revised the scheme following your request and now sample the middle of the liquid-dominated cloud top layer. The description of the methodology and Fig 2c is revised accordingly. Indeed, the results shown in Fig. 9 did not change significantly.

Minor comments:

Line 83: McFarquhar et al. paper is now published (2021), see <https://doi.org/10.1175/BAMS-D-20-0132.1>

R6: Replaced as suggested.

Table 2: I struggle to believe that Limassol's climate is 'northern tropics'. For a start, it's at 35N, well outside the tropical region. I suggest you change this phrase to more accurately reflect the climate zone Limassol is in.

R7: We changed it to Northern subtropics, which reflects the climate zone better.

Line 190: You use ECMWF IFS analysis for temperatures over Punta Arenas. The lack of radiosondes is somewhat unfortunate, but I fully agree with the authors in using an analysis product instead. So, some comment on the uncertainty in temperatures at cloud top height should be included though. While I doubt these uncertainties would often push your results into adjacent 5C-wide temperature bins, the

IFS temperatures will carry some uncertainties. Perhaps a Monte-Carlo simulation could be performed to test how a 1C or 2C uncertainty in cloud top temperature, of your thin stratiform clouds, changes the results. From my experience in comparing remote-sensing retrievals (radar and/or lidar), the (re)-analysis temperatures do not always match up with the melting level or cloud top inversion height.

R8: Thanks for sharing your experience. Referee #1 raised a related issue, please see R4.

Line 255 onwards: Can you add a figure, perhaps in an appendix, showing a typical example of clouds which are influenced by orographic waves please? It would be informative to visually see the differences as observed by your ground-based instruments.

R9: Thanks for the suggestion; we added an example case in the appendix (new Fig. A1).

Line 267: You need to explain why you select the 0.8 autocorrelation threshold. What happens when you vary this by e.g. changing to 0.7 or 0.9? How do the results change?

R10: The value of 0.8 is based on visual inspection of the whole dataset. When, changing the value at which the characteristic drop is estimated, the general results would not change, only the length of the shifts at which the autocorrelation threshold drops below the threshold value will change. For example, a value of 0.7 would require slightly larger shifts than 300 m to obtain a similar fraction of ice containing clouds. Defining a physically justified threshold would require a full characterization of the turbulent flow around the cloud. We included a brief discussion of this issue in the final paragraph of section 2.5. As also stated in response R11, we are working on a successor study about the properties of orographic waves.

Line 268 (approx.): The autocorrelation method is a good idea to determine the influence of orographic waves on cloud phase, and, as the authors describe, necessary at locations like Punta Arenas. To provide the broader context for wave forcing of clouds over Punta Arenas, and an additional verification on their method, I suggest that the authors composite the synoptic scale meteorology. I'd suggest using a reanalysis (ERA5?) at the closest time-step to the observed clouds, and compositing surface pressure and 10 m wind fields (as vectors) for the two cases of orographically-forced waves (autocorrelation>0.8, using your threshold in the paper) and no forcing (<0.8). In general, orographic waves form when relatively strong near-surface winds impinge upon a mountain range approximately perpendicularly (within 45degrees of this, e.g. see Section 5 in Dornbrack et al., 2001, JGR, <https://doi.org/10.1029/2000JD900194>). This is commonly observed in the Andes further north which are aligned north-south, but, around Punta Arenas, as the authors know, the topography is very complicated. The southern tip of the Andes is likely a unique region for cloud phase in the whole Southern mid-latitudes, so I think that understanding some of the synoptic features which generate the waves and result in enhanced amounts of liquid water would be useful for the community.

R11: Thank you for posing this interesting idea. Also from our current understanding, the occurrence of wave clouds is connected to specific meteorological conditions. But we have the feeling that incorporating the analysis of another dataset is beyond the scope of this study. We will definitely take a look at this topic in a follow-up study.

Figure 8: This is a nice, and informative, figure. However I wonder whether it's possible to increase readability, without losing the message, by plotting the average autocorrelations and PSDs in e.g. five degree temperature bands?

R12: Thank you for this comment. The curves of single clouds are now averaged into temperature bins of 5K.

Line 354: You could cite McFarquhar et al. (2021) here to support this point, who showed the measured INP values over the Southern Ocean and who also noted the ~3 orders of magnitude lower values than were reported in the 1970s.

R13: Done as suggested.

Line 402-403: You could expand your comments on Figure 7b as you only have 1 sentence at the moment.

R14: Thank you for the suggestion. We expanded the discussion of Fig. 7b.

Line 464: To support the statement that INPs in the BL are of biological origin over the Southern Ocean, how about citing Uetake et al. (2020) <https://doi.org/10.1073/pnas.2000134117>

R15: Added the reference. Thanks for the suggestion.

Technical corrections:

R16: Thank you for your technical suggestions. We implemented all of them as recommended.

Line 70: ‘...higher aerosol load allows us to advance...’

Line 84: ‘But, apart from the year-long...’

Line 103: ‘The goal of this...’

Line 113: ‘...and allow us to...’

Figure 2: Suggest you mention in the caption where the temperature is obtained from to avoid the reader having to dig through the text. You should also describe in the caption what the dashed line in Figure 2c is.

Line 241: ‘... horizontal extent of...’

Figure 5: Caption: ‘red squares in (c)...’

Line 380: ‘Limassol nearly all clouds...’

Line 380: Why not say ‘...fraction of (0.6 +/- 0.1) of shallow...’

Line 405: Well, ‘The lack of ice-containing cloud layers...’ seems pretty strong statement to me. Looking at Figure 7b, you still have them over Punta Arenas for about half of the clouds. Suggest rephrasing to ‘The reduced fraction of ice-containing clouds...’

Line 446: ‘absence of continental aerosol species...’

Line 453: ‘Murphy et al. (2019) argue in a ...’

Line 497: ‘CloudSat’

Line 520: ‘... with orographic gravity waves...’

Figure A1 & A2: Add units to x-axes

R17: The unit of the normalized residence time is [s s⁻¹]. Thus we omitted it.

Further changes

A few changes were incorporated due to additional feedback.

1. The discussion of the Cotton and Field 2002 paper was misleading. We clarified that sentence (L244f original draft).
2. The caption of Tab. 3 was slightly expanded and the ice extinction, later used for the lidar detection threshold, was added.
3. The instrument naming in Tab. 1 was slightly refined.
4. Replaced 'cloud object' with 'cloud case' and 'land cover' with 'surface cover' throughout the manuscript.
5. We refined a few formulations throughout the manuscript, to make them clearer without changing the meaning.

Hemispheric contrasts in ice formation in stratiform mixed-phase clouds: Disentangling the role of aerosol and dynamics with ground-based remote sensing

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Abstract.

Multi-year ground-based remote-sensing datasets were acquired with the Leipzig Aerosol and Cloud Remote Observations System (LACROS) at three sites: ~~a~~. A highly polluted central European site (Leipzig, Germany), a polluted and strongly dust-influenced eastern Mediterranean site (Limassol, Cyprus), and a clean marine site in the southern mid-latitudes (Punta Arenas, Chile) are used to contrast ice formation in shallow stratiform liquid clouds. These unique, long-term datasets at key sites in key regions of aerosol-cloud interaction provide a deeper insight into cloud microphysics. The influence of temperature, aerosol load, boundary-layer coupling, and gravity wave motion on ice formation is investigated. With respect to previous studies of regional contrasts in the properties of mixed-phase clouds our study contributes the following new aspects: (1) Sampling aerosol optical parameters as a function of temperature, the average backscatter coefficient at supercooled temperatures-conditions is within a factor of 3 at all three sites. (2) Ice formation was found to be more frequent for cloud layers with cloud top temperatures above -15°C than indicated by prior lidar-only studies at all sites. A virtual lidar-detection threshold of IWC needs to be considered in order to bring radar-lidar-based studies in agreement with lidar-only studies. (3) At similar temperatures, cloud layers which are coupled to the aerosol-laden boundary layer show more intense ice formation than de-coupled clouds. (4) Liquid layers formed by gravity waves were found to bias the phase occurrence statistics below -15°C . By applying a novel gravity wave detection approach using vertical velocity observations within the liquid-dominated cloud top, wave clouds can be classified and excluded from the statistics. After considering boundary layer and gravity-wave influences, Punta Arenas shows lower fractions of ice containing clouds by 0.1 to 0.4 absolute difference at temperatures between -24 and -8°C . These differences are potentially caused by the contrast in the INP reservoir between the different sites.

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20 1 Introduction

Clouds and aerosol are inseparably ~~coupled,~~ linked via complex pathways of interaction whose outcome manifests in the macroscopic properties of precipitation and radiation fields. ~~On the one hand~~In the first place, aerosol particles are required as cloud condensation nuclei (CCN) on which cloud droplets can form. On the other hand, primary ice formation in the heterogeneous freezing temperature range from 0 to ~~approximately about~~ -40°C requires ice nucleating particles (INP) to
25 be present in the aerosol reservoir. The ways in which aerosol and cloud particles interact are controlled by the dynamics and thermodynamics of the atmospheric environment, with temperature being the most important driver. Thermodynamic processes are considered to dominate the cloud microphysical properties ~~because they over aerosol-related processes because~~
the thermodynamics control the amount of water ~~vapour~~ vapor that is available for being transferred to either the liquid or the ice phase. This dominance makes it difficult to isolate aerosol-related effects in observations of cloud properties.

30 ~~Nevertheless~~Despite the dominance of dynamics and thermodynamics, observations as well as aerosol-permitting model studies suggest a considerable influence of the aerosol conditions on the properties and evolution of clouds and precipitation (Seifert et al., 2012; Possner et al., 2017; Solomon et al., 2018; Zhang et al., 2018). Solomon et al. (2018) used high-resolution modelling of Arctic mixed-phase clouds to show ~~,~~ that perturbations in the INP concentration dominate over changes in the ~~cloud condensation nuclei (CCN)~~ CCN concentrations. Cloud chamber studies suggest ~~,~~ that holding CCN constant, the ratio
35 of ice to liquid water content in the steady state is predominantly controlled by INP concentrations (Desai et al., 2019).

There is also a distinct spatio-temporal variability of the performance of weather and climate model simulations, which is attributed to the insufficient representation of aerosol-cloud-dynamics interaction ~~processes~~ in the models (Fan et al., 2016; Seinfeld et al., 2016). For instance, ~~the reasons for the a~~ less accurate treatment of the radiative balance in the southern-hemispheric mid-latitudes compared to their northern-hemispheric ~~counterpart are still debated~~ counterparts was found (Tren-
40 berth and Fasullo, 2010; Grise et al., 2015). The atmosphere of the southern hemisphere's mid-latitudes is a unique component of the Earth's climate system. It's the stormiest ~~(e.g. Young, 1999)~~ (e.g., Young, 1999) and one of the cloudiest places on Earth (Haynes et al., 2011; Naud et al., 2014), but process understanding of clouds in that region is still limited. The reported biases in the solar radiation budget are attributed to shallow supercooled ~~liquid topped~~ liquid topped clouds, which are insufficiently represented by current models (Bodas-Salcedo et al., 2014; Kay et al., 2016; Bodas-Salcedo et al., 2016; Kuma et al., 2020).
45 ~~These radiation biases affect estimates of sea surface temperature and surface precipitation, ultimately affecting the energy balance~~ With the frequency of liquid-cloud tops too low, less short-wave radiation is reflected. Hence, the simulations show a too strong heating at the surface, leading to a bias in ocean heat uptake (Franklin et al., 2013; Hyder et al., 2018). There is an ongoing controversy about the reasons for the observed differences and prevailing model deficiencies, but indications are given that a combination of hemispheric contrasts in aerosol load and atmospheric dynamics plays a role.

50 Different causes for ~~the excess of the~~ regional contrasts in the abundance of supercooled liquid cloud layers are proposed. On the one hand, reason for the excess of supercooled liquid water in southern-hemispheric cloud systems could be the inhibition of ice formation caused by the lack of ice nucleating particles in the predominantly pristine environment of the Southern Ocean (Hamilton et al., 2014), where terrestrial sources, which are frequently considered a good source for INP, are rare or

far apart (Vergara-Temprado et al., 2017). In the heterogeneous freezing regime, suitable aerosol particles are a prerequisite
55 for ~~ice formation and missing the formation of ice~~. Missing ice formation as a sink for cloud water, the liquid phase may be sustained for long periods of time. On the other hand, dynamical processes ~~could~~ can lead to an enhancement of supercooled liquid water. Korolev (2007) showed ~~that~~ depending on the number and size of ice crystals, a threshold vertical velocity can be found, which allows for sufficient supersaturation to grow the ice as well as the liquid phase. Gravity waves have been suggested to play a role in the phase partitioning of Southern Ocean clouds (Alexander et al., 2017; Silber et al., 2020). Due
60 to the orographic effects and the strong westerlies, gravity waves are a general feature in the vicinity of all landmasses in the middle and high latitudes of the southern hemisphere (Sato et al., 2012; Alexander et al., 2016). It is however also noteworthy that stronger turbulence increases the amount of ice formed in stratiform cloud layers (Bühl et al., 2019). Indications are thus given that it is necessary to also consider turbulence in studies of ice formation.

The undetermined contributions of dynamical and aerosol effects on the observed excess of supercooled liquid in the south-
65 ern hemisphere requires dedicated ~~attribution~~ contrasting studies. In numerous previous studies, liquid-topped supercooled stratiform cloud layers have been proven to be suitable natural laboratories for the investigation of the relationships between aerosol properties, thermodynamics, and microphysical properties of clouds in the heterogeneous freezing regime. Temperature at which the ice formation occurs, needs to be strongly constrained, because the concentration of efficient INP increases rapidly with decreasing temperature (~~e.g. Kanji et al., 2017~~) (e.g., Kanji et al., 2017). Accordingly, the amount of ice formed
70 also increases orders of magnitude for lower temperatures (Bühl et al., 2016). The lowest temperature in such clouds occurs on top of the liquid-dominated layer, hence the cloud top temperature (CTT) can be used to constrain the ice formation temperature. Turbulence is usually confined to the liquid-dominated cloud top (Westbrook and Illingworth, 2013; de Boer et al., 2009) and due to the limited thickness of this layer, secondary ice formation or ice multiplication are strongly constrained (Fukuta and Takahashi, 1999; Myagkov et al., 2016). Contrasting microphysical properties observed in pristine, clean regions with
75 observations from areas with higher aerosol load ~~e.g., allows~~ allows us to advance understanding of the impact of different aerosol loads (Choi et al., 2010; Kanitz et al., 2011; Seifert et al., 2015; Tan et al., 2014). Recent studies based on the A-Train satellite constellation suggest systematically lower ice amounts in the southern mid-latitudes (Zhang et al., 2018) and a strong susceptibility to dust load (Villanueva et al., 2020). Supercooled liquid clouds are frequent over the Southern Ocean (Huang et al., 2015; Hu et al., 2010) and studies by Kanitz et al. (2011) and Choi et al. (2010) showed ~~that at similar temperatures~~
80 ~~ice that at similar temperatures ice~~ is formed less frequently by liquid layers in the southern hemisphere mid-latitudes, than in the northern hemisphere. The study of Kanitz et al. (2011) first used a ground-based lidar at Punta Arenas (53.1°S 70.9°W, Chile) to assess the thermodynamic phase of stratiform mixed phase clouds above the Southern Hemisphere mid-latitudes. Major caveats of this study are the limited duration of the observations during austral summer and the limitations of the lidar-only setup. Alexander and Protat (2018), using ground-based lidar and A-Train observations from Cape Grim (40.7°S 144.7°E, Aus-
85 tralia) confirm the basic findings also for the eastern parts of the Southern Ocean. ~~This~~ Their study, similarly to McErlich et al. (2021), emphasizes the problems in A-Train derived datasets detecting shallow clouds in the lowermost part of the atmosphere.

Recent activities include shipborne, land-based and aircraft campaigns targeting aerosols and clouds above the Southern Ocean between Australia and Antarctica (60 to 160°E). An overview is provided by ~~?~~ McFarquhar et al. (2021). In terms of

stratiform clouds, the large abundance of supercooled liquid water, occasionally down to -30°C , was confirmed. But, apart from the year-long lidar/radar dataset at Macquarie Island (54.6°S 158.9°E , Australia) the cloud observations focused on austral summer.

Using a shipborne dataset, Mace and Protat (2018) also found frequent liquid-dominated clouds with low radar reflectivities and ~~1/3~~ one-third of the liquid layers only observed with lidar. Comparing the observations with a Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO) dataset from Hu et al. (2010), they found an overestimation of supercooled liquid in the satellite dataset, especially strong at temperatures above -15°C . In a follow-up study, Mace et al. (2020) refined the CALIPSO classification scheme, leading to more frequent detections of the mixed phase, especially during wintertime and in the lower latitudes of the Southern Ocean. However, no CTT-resolved phase occurrence statistics is presented. Liquid layers in deeper clouds, observed during another shipborne campaign (~~?~~Alexander et al., 2021) (McFarquhar et al., 2021; Alexander et al., 2021), could only be reproduced in regional model simulations, when INP parametrization was tuned to lower concentrations (Vignon et al., 2021). Zaremba et al. (2020) investigated airborne active ~~remote-sensing~~ remote-sensing observations of Southern Ocean clouds south of Tasmania. They also found widespread liquid cloud tops at temperatures down to -30°C . By investigating the ground-based ~~remote-sensing~~ remote-sensing dataset assembled at McMurdo (77.8°S 166.7°E , Antarctica), Silber et al. (2018) found frequent long-lived ~~liquid-topped~~ liquid-topped clouds, also below -30°C .

Yet, a statistical analysis of the relationship between both, aerosol conditions, cloud vertical dynamics, and the phase partitioning in stratiform cloud layers of the southern-hemisphere mid-latitudes based on long-term observations was not established. One reason is ~~;~~ that, despite increased activity in the recent past, ground-based ~~remote-sensing~~ remote-sensing observations of clouds and aerosol are still sparsely distributed in the Southern Ocean and at the coast of Antarctica.

~~Goal-~~The goal of this study is to analyze long-term ground-based ~~remote-sensing~~ remote-sensing observations of aerosol properties, cloud microphysics, and atmospheric dynamics from three sites with strongly contrasting aerosol conditions in order to attribute ice formation in the heterogeneous freezing regime to atmospheric dynamics and aerosol conditions. For that ~~attribution-contrasting~~ approach, we utilized the recent campaigns of the Leipzig Aerosol and Cloud Remote Observations System (LACROS) at Leipzig (51.4°N 12.4°E , Germany), Limassol (34.7°N 33.0°E , Cyprus), and Punta Arenas (53.1°S 70.9°W , Chile), which provide datasets ~~;~~ that cover the aerosol conditions of a continental northern hemispheric background site, a hot-spot of mineral dust, and the marine-dominated pristine Southern Ocean, respectively. Hence, these datasets collected with a single set of ground-based ~~remote-sensing~~ remote-sensing instrumentation provide an ideal basis for contrasting studies. The broad variety of instruments covers the decisive properties of aerosols, dynamics, clouds, and precipitation for a more comprehensive picture of aerosol-cloud interaction. The observations at Punta Arenas provide the first multi-year dataset of synergistic ground-based remote sensing observations in the western half of the Southern Ocean and ~~allow~~ allows us to contextualize prior findings.

The paper is structured as follows: ~~In section 2 the instrumentation and~~ The instruments and field campaigns are described first (Sec. 2.1), followed by the retrievals of aerosol properties (Sec. 2.2) ~~and the synergistic Cloudnet retrieval (Sec. 2.3).~~ Afterwards, the methods for cloud selection and vertical velocity characterization are introduced. Instruments, field

campaigns and synergistic retrievals are described first. Section 2 covers the methods, consisting of the retrieval of the aerosol statistics (2.2), the automated cloud selection algorithm (Sec. 2.4) (2.4), and the characterization of vertical velocity (Sec. 2.5). The statistics is results are presented in Sec. 3, including the lidar-derived average profiles of aerosol optical properties (Sec. 3.1) and the cloud phase (3.1), and frequency of ice-formation statistics with special emphasis on instrument sensitivity (Sec. 3.2.1). Boundary layer coupling (Sec. 3.2.2) and vertical dynamics are discussed in Sec. 3.2.3 3.2.2 and Sec. 3.2.3, respectively. The amount and efficiency of ice production is assessed in Sec. 3.2.4. The study concludes with a discussion of the contrast identified in aerosol load and stratiform cloud properties (Sec. 4) followed by a summary and outlook (Sec. 5).

2 Data and Methods

This section introduces the datasets and methods used in the remainder of this study. Starting with the campaigns and instrumentation (Sec. 2.1), followed by a short description of the retrievals used (Sec. 2.2 and 2.2 and 2.3) and finally the methods for automated selection of shallow stratiform clouds (Sec. 2.4) and characterization of vertical velocity dynamics motion (Sec. 2.5).

2.1 Datasets from Leipzig, Limassol, and Punta Arenas

Basis of the observational datasets presented below is the Leipzig Aerosol and Cloud Remote Observations System (LACROS), the mobile ground-based remote-sensing supersite of the Leibniz Institute for Tropospheric Research (TROPOS), Leipzig, Germany. The instrumentation used for the synergistic approaches applied in this study comprises a Mira-35 MIRA-35 35 GHz scanning cloud radar, a Polly^{XT} multi-wavelength Raman and depolarization polarization lidar, a Streamline XR 1.5 μ m scanning Doppler lidar, a HATPRO 14-channel microwave radiometer, a 1064 nm ceilometer, an optical disdrometer, and radiation sensors. Main properties of the sensors are summarized in Table 1.

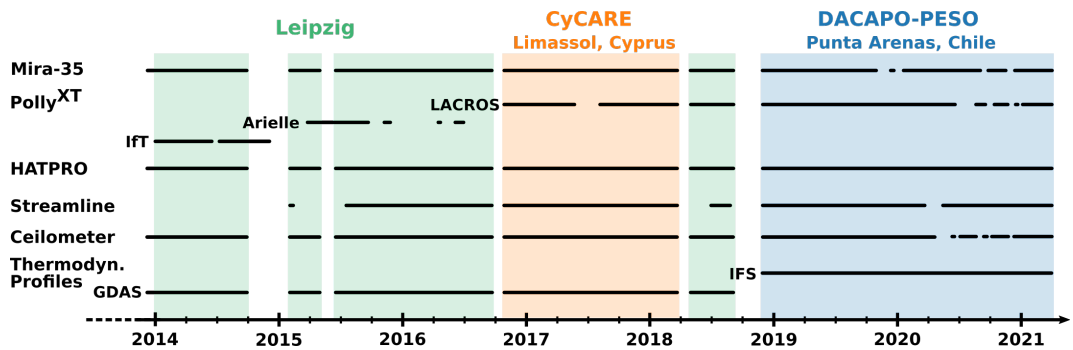


Figure 1. Overview on instrument availability and campaign durations of the LACROS deployments covered in this study. Characteristics of the three different Polly^{XT} instruments used at Leipzig and named IFT, Arielle, and LACROS are described in Engelmann et al. (2016).

LACROS was established in 2011 and was first introduced by Bühl et al. (2013b). After the initial setup phase, the basic set of instrumentation of LACROS has been kept unchanged since the year 2014. Hence, this comparative study uses data from the

Table 1. Specifications of the LACROS instruments [and measured quantities](#) used in this study.

Instrument (Reference)	Frequency ν Wavelength λ	Measured quantity	Temporal resolution	Vertical range	Vertical resolution
Doppler cloud radar METEK MIRA-35 (Görsdorf et al., 2015)	$\nu = 35$ GHz	Radar reflectivity factor	3.5 s	150 – 13000 m	30 m
		Vertical velocity	3.5 s	150 – 13000 m	30 m
		Linear depolarization ratio	3.5 s	150 – 13000 m	30 m
Raman-Polarization Lidar Polly ^{XT} (Engelmann et al., 2016)	$\lambda = 355, 532, 1064$ nm	Attenuated backscatter coeff.	30 s	100 – 15000 m	7.5 m
	$\lambda = 355, 532$ nm	Raman backscatter signal	1 h	300 – 5000 m	~ 50 m
	$\lambda = 355, 532$ nm	Linear depolarization ratio	30 s	100 – 15000 m	7.5 m
Microwave radiometer RPG HATPRO-G2 (Rose et al., 2005)	$\nu = 22.24 - 31.4$ GHz	Brightness temperatures	1 s	column integral	
	$\nu = 51.0 - 58.0$ GHz	Brightness temperatures	1 s	column integral	
Doppler Lidar HALO Photonics Streamline XR (Pearson et al., 2009)	$\lambda = 1.5$ μ m	Attenuated backscatter coeff.	2 s	48 – 12000 m	48 m
		Vertical velocity	2 s	48 – 12000 m	48 m
Ceilometer Jenoptik ehm15kx CHM15kx	$\lambda = 1064$ nm	Attenuated backscatter coeff.	30 s	15 – 15300 m	15 m
Optical disdrometer Ott Parsivel ² (Löffler-Mang and Joss, 2000)	$\lambda = 650$ nm	Hydrometeor size distribution	30 s	4 m	-
Sun photometer Cimel CE318-T (Barreto et al., 2016)	$\lambda = 340 - 1064$ nm	Aerosol optical thickness	variable	column integral	

6 year period since then. During that period, LACROS was, besides several 4-8 weeks short-term deployments at other sites,
145 stationed at Leipzig, Limassol, and Punta Arenas.

Fig. 1 provides an overview on the campaigns and instrument availability. From October 2016 to March 2018 LACROS
was located at Limassol for the Cyprus Clouds, Aerosol, and Rain Experiment (CyCARE). In the context of the present
study, this location in the eastern Mediterranean in the middle of the dust belt serves as a reference for a highly aerosol-
laden environment. First results of the CyCARE campaign are described by Ansmann et al. (2019). For the second long term
150 campaign, LACROS was ~~moved to Punta Arenas in November 2018~~ [deployed at Punta Arenas](#) for the two-year Dynamics,
Aerosol, Clouds, And Precipitation Observations in the Pristine Environment of the Southern Ocean (DACAPO-PESO) field
campaign ~~since November 2018~~. Located at 53.1°S, Punta Arenas is in the midst of the Southern Ocean, farther south than
any other continental land mass or major island apart Antarctica. Under prevailing westerly flow, the next land mass upwind
is more than 8000 km away and significantly to the north. Hence, the free tropospheric aerosol is predominantly of marine
155 origin with low concentrations overall. First results of the DACAPO-PESO campaign regarding events of significant aerosol
load (Ohneiser et al., 2020; Floutsi et al., 2021), liquid cloud microphysics (Jimenez et al., 2020) and integrated water vapor
observations (Bromwich et al., 2020) are already published. Observations at Leipzig were performed before October 2016 and

Table 2. Overview on Location, data availability, climate, aerosol load, and related studies for the LACROS datasets used.

Location	Leipzig, Germany	Limassol, Cyprus	Punta Arenas, Chile
	51.4°N 12.4°E	34.7°N 33.0°E	53.1°S 70.9°W
Station altitude	125 masl	11 masl	9 masl
Campaign name		CyCARE	DACAPO-PESO
Duration	976 d	524 d	765 d
Cloudnet data	771 d	520 d	674 d
Doppler lidar data	513 d	523 d	717 d
Polly ^{XT} data	633 d	460 d	702 d
Climate	Northern mid-latitudes	Northern Tropics <u>subtropics</u>	Southern mid-latitudes
Typical aerosol load	Continental background, occasionally dust	Dust, marine, continental	Marine, occasionally continental
Related studies	Bühl et al. (2016) <u>Ansmann (2005)</u> <u>Seifert et al. (2010)</u> <u>Bühl et al. (2013a)</u> <u>Bühl et al. (2016)</u>	Ansmann et al. (2019)	Ohneiser et al. (2020) Bromwich et al. (2020) Jimenez et al. (2020) Floutsi et al. (2021)

between the deployments to Limassol and Punta Arenas. Due to the location in central Europe, the Leipzig dataset serves as a reference for continental background in the northern hemisphere.

160 Data coverage for each of the sites is summarized in Table 2. Gaps in the observations were usually caused by periods of short-term deployments at other sites, failure of primary instruments, or power cuts (see also Fig. 1). Coverage with the core instrumentation is generally well above 80%, only exception being Doppler lidar and Polly^{XT} at Leipzig.

2.2 Lidar-based aerosol statistics

As a proxy for ~~assessing~~ the aerosol conditions at the three measurement sites, we used vertically resolved observations from
165 the Polly^{XT} lidar system (Althausen et al., 2009; Engelmann et al., 2016). Quantities of interest are the aerosol backscatter coefficient, the extinction coefficient, the particle linear depolarization ratio, and the cloud-relevant concentration of INP. Their retrieval is explained below.

Basis for the statistical analysis are profiles of particle backscatter coefficient β_p at 532 nm wavelength. The profiles are computed with the Klett method (Fernald, 1984) by the PollyNET ~~retrievals whenever~~ retrieval whenever atmospheric
170 conditions are suitable (Baars et al., 2016, 2017; Yin and Baars, 2021). The PollyNET retrieval chain also ensures a homogenized analysis of the data from the three different Polly^{XT} instruments, which were utilized in the frame of this study (~~see Fig. 1 and Engelmann et al., 2016; Baars et al., 2016~~)(see Fig. 1 and Engelmann et al., 2016; Baars et al., 2016).

Profiles of the particle linear depolarization ratio (hereafter referred to as particle depolarization ratio) are only calculated when the ratio of molecular backscatter coefficient to β_p ~~exceeds~~ is below a value of 18. Additionally, any particle depolar-
175 ization ratios larger than 0.7 are masked, as they are indications for noise artifacts in the cross-polarized signal component.

All profiles are then filtered with the co-located Cloudnet target classification (see Section 2.3) to exclude clouds, especially optically thin ice clouds ~~only seen that are only clearly classified~~ by the cloud radar. Finally, a manual screening excluded fragments of thin liquid clouds, which would otherwise artificially increase β_p . For the averages, the optical data of each retrieved profile is binned to vertical intervals of 200 m or 3 K.

180 The derived average optical properties can be used to estimate aerosol microphysical properties, such as concentrations of ice nucleating particles (INP). This is an important step in order to evaluate the datasets of the three sites with respect to contrasts in the potential contribution of aerosol effects on heterogeneous ~~ice formation~~ ice formation efficiency. Conversion from optical properties as observed by lidar to microphysical aerosol properties is based on the parametrizations described by Mamouri and Ansmann (2016). By means of this approach, the lidar-measured aerosol extinction coefficient is converted to
185 the number and surface concentration N_{500} and S_{500} ~~respectively~~ of aerosol particles larger than 500 nm in diameter. These quantities are applied in available in-situ-based parametrizations for the retrieval of INP concentrations. Prerequisite for the retrieval is a correct aerosol typing, as different types of particles differ by orders of magnitude in their ice forming efficiency. In order to do so, the average backscatter profile is separated into the categories marine, continental, and mineral dust, based on air mass source ~~(see Appendix B and Radenz et al., 2021)~~ (see Appendix B and Radenz et al., 2021) and particle depolar-
190 ization ratio (one-step POLIPHON; Mamouri and Ansmann, 2017). The average extinction is calculated from the profiles of β_p by assuming a typical lidar ratio ~~marine of 20 sr, continental for marine, 50 sr and dust for continental, and 45 sr for dust~~ aerosol (Müller et al., 2007; Baars et al., 2017; Bohlmann et al., 2018). ~~The In a next step, the~~ extinction coefficient is ~~in the next step~~ converted to N_{500} and S_{500} , using sun-photometer-based conversion factors (Mamouri and Ansmann, 2016). Then, the above mentioned INP parametrizations are applied for each aerosol class, such as DeMott et al. (2015) for mineral dust,
195 DeMott et al. (2010) for continental aerosol ~~or~~ and McCluskey et al. (2018b, a) for marine aerosol.

2.3 Cloudnet processing

~~For Synergies between lidar, cloud radar, microwave radiometer, and meteorological data are utilized for~~ determination of cloud macro- and microphysical properties and as basis for the stratiform cloud identification ~~synergies between lidar, cloud radar, microwave radiometer and meteorological data are utilized~~ scheme. State-of-the-art routines for achieving this ~~requirement~~
200 synergy are comprised in the Cloudnet retrieval (Illingworth et al., 2007). Cloudnet re-grids the observations to a common resolution (30 s and 31.18 m, determined by the vertical resolution of ~~Mira-35~~ MIRA-35 with a pulse length of 208 ns) and provides products, such as a target classification and ~~retrievals of~~ microphysical cloud properties. Regridding is a crucial step, as each of the instruments has different temporal and vertical resolution (Tab. 1). Regular scans of the MIRA-35 cloud radar (hourly) and the Streamline Doppler lidar (twice per hour) are not used within the Cloudnet processing scheme. Profiles of
205 temperature, pressure, and humidity are obtained from the ECMWF's IFS analysis for Punta Arenas and GDAS analysis for Leipzig and Limassol. Liquid water path (LWP) and integrated water vapor (IWV) are retrieved from brightness temperature observations of the microwave radiometer in two frequency bands from 22.24 to 31.4 GHz and 51.0 to 58.0 GHz ~~(7 channels each)~~ (7 channels each; Crewell and Löhnert, 2007). The statistical retrieval is based on long-term radiosonde observations (Leipzig and Limassol) and high-resolution reanalysis data (Punta Arenas). Attenuated backscatter of the ceilometer is

210 regularly cross-calibrated with Polly^{XT} using the calibrated attenuated backscatter of PollyNET. ~~Gaps in~~ Usually, the ceilometer data is used in the synergistic retrieval, as the dataset is more robust and less prone to interruptions. Rare gaps in the observations are filled with the Polly^{XT} ~~observations~~ attenuated backscatter at 1064 nm.

2.4 Automated cloud selection and characterization

~~While Cloudnet provides a concise overview on the macrophysical properties of the observed cloud layers, it requires dedicated, automatic and reproducible filtering algorithms~~ While the Cloudnet algorithm provides a pixel-by-pixel classification of cloud phase and microphysical properties, information on temporal coherence and cloud evolution is not readily available. Hence, an automatic reproducible filtering algorithm for the selection of ~~the~~ targeted stratiform, supercooled cloud systems ~~is necessary~~. Based on the data cube LARDA³ (Bühl et al., 2021), the approach of Bühl et al. (2016) is implemented into an automated selection algorithm.

220 An example of the Cloudnet processing of measurement data and the application of the cloud selection scheme is shown in Fig. 2. Starting with a profile of the Cloudnet target classification mask (Hogan and O'Connor, 2004), consecutive pixels classified as cloud pixels (liquid droplets, ice, ice and supercooled droplets) are grouped together and defined as features. ~~Then~~ In case similar types of hydrometeors were observed in matching heights, single features in neighboring ~~profiles~~ timesteps are connected to coherent cloud ~~objects, if similar types of hydrometeors were observed in matching heights cases~~. For the analysis, the cloud ~~objects~~ cases are filtered for shallow stratiform clouds, which are liquid topped and either have an ice virga or not (rectangles in Fig. 2d). An overview of the microphysical parameters sampled for each cloud ~~object~~ case is provided in Table 3. It is assumed ~~that~~ if ice is formed in a liquid layer it also sediments out of the cloud. This is required, as the signal in the top layer is dominated by return from liquid droplets and Cloudnet provides no reliable mixed-phase classification there.

To pinpoint potential effects of aerosol load, thermodynamic, and dynamic drivers of ice formation have to be constrained. 230 This is especially important, as the sites are situated in different climate zones. We presume for our study ~~that~~ thin stratiform clouds serve as a natural laboratory with only a limited number of microphysical processes being possible.

~~Clouds driven by convective processes are excluded by only allowing clouds that were observed for~~ The cloud cases are filtered for a length of more than 20 minutes and ~~showed a~~ smooth cloud top height ~~heights~~ (standard deviation < 150 m) ~~to exclude convective clouds. Laboratory studies of Fukuta and Takahashi (1999) and the cloud radar observations by~~ Myagkov et al. (2016) indicate that the average thickness of the liquid dominated cloud top layer has to be less than 350 m ~~This criterion is motivated by Fukuta and Takahashi (1999) and excludes in order to restrict the dataset to the regime of pristine ice formation and avoid strong effects of~~ secondary ice formation ~~processes and riming (see also Myagkov et al., 2016)~~ ~~Seeding by ice clouds above is avoided by excluding clouds with ice pixel above the liquid layer. Only cloud~~ objects cases with top temperatures between -38 and 0°C are considered for the statistics. For the phase occurrence frequency statis- 240 tics, a cloud is classified as ice producing, if ice pixel were observed 180 m below the liquid-dominated layer within at least 5% of the duration. On the other hand, cloud ~~objects~~ cases are classified as liquid-only, if ice pixel are observed to sediment out of the liquid-dominated layer less than 5% of the time. This threshold is simpler, than the decision tree used by Bühl et al. (2013a), but provides similar results. The resulting fraction is weighted by the length of the cloud. Similarly to

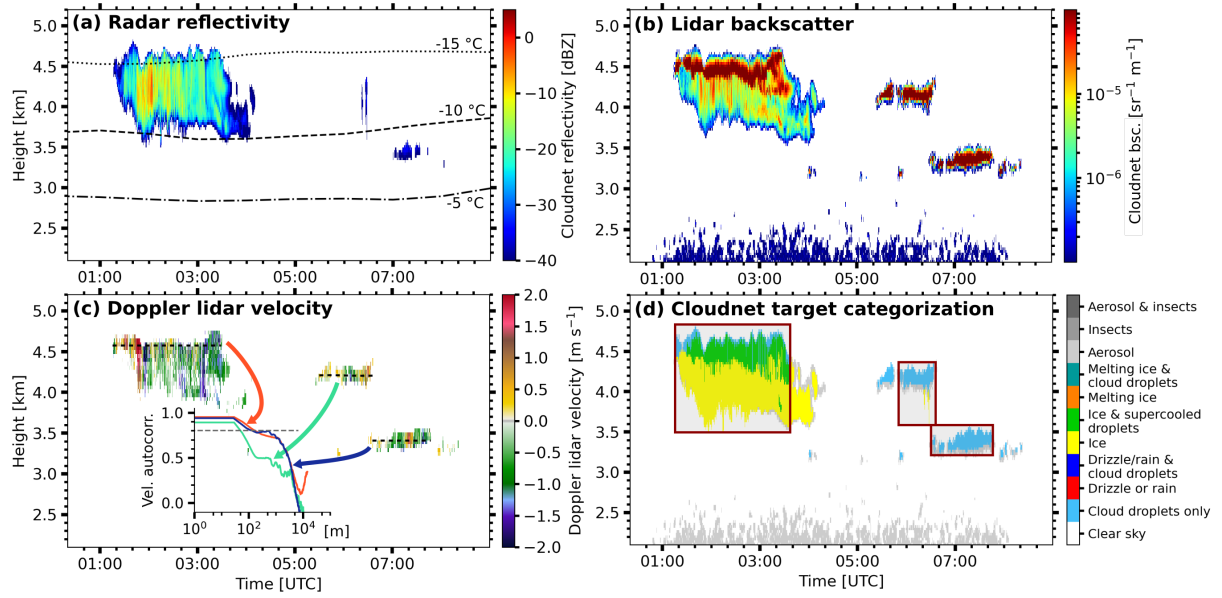


Figure 2. MIRA-35 Stratiform liquid-topped clouds as observed with LACROS. (a) Cloud radar reflectivity overlaid with the temperature, (ab) PollyXT Lidar attenuated backscatter, (bc) Streamline Doppler lidar vertical velocity(e), and (d) Cloudnet target categorization (d) on the 28 November 2018 between 00:20 and to 09:00 UTC. The temperature information in (a) is obtained from ECMWF IFS model data. Red rectangles in (d) show indicate the automatically detected liquid-topped shallow stratiform clouds. The inset in (c) shows the autocorrelation function of the vertical velocities in the liquid-dominated layer (heights indicated by dashed lines) of each cloud. The height of the velocity sampling is indicated with dashed lines. Further details details are provided in Sec. 2.5.

(Bühl et al., 2016) Bühl et al. (2016), the properties of the ice in the virga are taken 180 m below the liquid-dominated cloud top layer to avoid uncertainties in the liquid base estimate and sublimation within the virga. The ice water content (IWC) is derived from the radar reflectivity and the temperature using the Hogan et al. (2006) retrieval. With this automated cloud selection scheme multi-year datasets can be analyzed using objective criteria, while yielding statistics similar to manual cloud selection (e.g. Seifert et al., 2010; Kanitz et al., 2011; Seifert et al., 2015) (e.g., Seifert et al., 2010; Kanitz et al., 2011; Seifert et al., 2015)

250 2.5 Gravity wave detection

In order to enable the attribution pinpoint effects of aerosol and dynamical effects dynamics on the phase partitioning in the stratiform cloud dataset, an approach is required to assign cloud dynamics vertical motion regimes to each cloud object case. Here we focus on the temporal structure of vertical velocity, to constrain the dynamics forcing on a cloud. Usually, shallow clouds are characterized by a fully developed turbulence in the liquid-dominated cloud top (Bühl et al., 2019), where the vertical motion is driven by cloud top cooling (e.g. Shao et al., 1997; Fang et al., 2014; Simmel et al., 2015)

Table 3. Parameters sampled from the atuomatically identified cloud objectscases. A more detailed description of the automated spatio-temporal selection method is provided in Bühl et al. (2016). The IWC and ice extinction is retrieved with the parametrization of Hogan et al. (2006).

Parameter	Time series	Description
Cloud top temperature	yes	Temperature of the topmost liquid pixel
Cloud top height	yes	Geometrical height of the topmost liquid pixel
Liquid layer thickness	yes	Geometrical depth of the liquid-dominated cloud top layer
Phase classification	no	Ice pixel observed below liquid-dominated layer > 5% of duration
Cloud-base <u>In-cloud</u> vertical velocity	yes	Doppler lidar vertical velocity at the base of the liquid-dominated <u>in the middle of the cloud top</u>
Liquid-water content <u>LWC</u>	yes	Liquid-water content <u>LWC</u> of the liquid-dominated layer
Reflectivity virga	yes	Radar reflectivity 180m below the base of the liquid-dominated layer
Ice-water content <u>IWC</u> in virga	yes	Reflectivity-derived <u>Reflectivity-derived</u> IWC 180m below the base of the liquid-dominated layer
<u>Extinction in virga</u>	<u>yes</u>	<u>Optical extinction of ice at 180m below the base of the liquid-dominated layer</u>
Ice-to-liquid content ratio	yes	Ratio of IWC virga to LWC <u>in the liquid-dominated layer</u>

(e.g., Shao et al., 1997; Fang et al., 2014; Simmel et al., 2015). In the turbulent layer at cloud top, up- and downdrafts alternate at horizontal scales in the order of 100 m or less.

However, orographic gravity waves can trigger vertical motion and associated cloud formation, as well. Microphysical processes in these wave clouds are governed by large-scale dynamics, where vigorous up- and downdrafts may appear stationary.

260 Due to this dynamics, the mixed-phase and the ice phase are horizontally separated, with the liquid drops predominantly in the ascending branch and the ice particles in the descending branch (Heymsfield and Miloshevich, 1993; Baker and Lawson, 2006). The properties of the horizontal wind field determines the regions of the up- and downdraft in such orographic clouds. Observing these clouds by stationary ground-based remote sensing might thus not sample the full horizontal ~~extend~~extent of the cloud, which causes a misclassification of the cloud case in terms of liquid-only and ice-producing. An illustrative example

265 of such a wave cloud is shown in appendix A

Also, the flow is highly laminar, opposed to the confined, fully developed turbulence found in layered mixed-phase clouds. Cloud microphysics in wave conditions cannot directly be compared to layered clouds. ~~Ice formation in those clouds is dominated by homogeneous and evaporation freezing (Cotton and Field, 2002), which are not subject to this study and should be excluded from our statistics.~~ For clouds in the heterogeneous freezing regime, Cotton and Field (2002) found that only rapid

270 evaporation freezing once the downdraft commences could explain their observations, where evaporation freezing can be better characterized as inside-out contact freezing of shrinking particles (Durant, 2005). A more recent study by Field et al. (2012) found that condensation and immersion freezing are needed together with deposition and evaporation freezing to explain their aircraft observations of ice formation in wave clouds.

On the other hand, the frequent occurrence of atmospheric gravity waves in a specific region might increase the frequency

275 of thermodynamic conditions that favor the presence of a sustained liquid phase. As Korolev (2007) demonstrate, long-lasting

steady updrafts are required in order to make the liquid phase dominating over the ice phase. Probing an observational dataset for the presence of long-lasting updrafts could therefore provide a hint on the role of atmospheric gravity waves in the occurrence of enhanced concentration of supercooled liquid.

280 Turbulence properties of the liquid layer in the clouds subject to our study can be derived from the vertical velocity observation by Doppler lidar (Bühl et al., 2019). The small size of droplets in the liquid-dominated cloud-top layer and their negligible terminal velocity makes them tracers of air motion. From the Doppler lidar observations, the vertical velocity is sampled at the ~~pixel with the maximum backscatter out of the heights~~ average height of the center of the layer identified as liquid-containing in the Cloudnet classification. The height of this sampling is indicated as dashed lines in Fig. 2e 2c and Fig. A1d. The temporal resolution of the resulting time series is 2 s, ~~equally i.e., equal~~ to the Doppler lidar raw data. This time-series is then used to get
285 insights into the turbulent properties of the ~~cloud-top layer.~~

~~The structure of the vertical velocity timeseries can be characterized with the autocorrelation function~~ cloud-top layer. To separate both regimes in the large dataset, an characteristic parameter is needed. The autocorrelation function was found to be a pragmatic choice for such a parameter.

The autocorrelation function $\Psi(\tau)$ for a time series of vertical velocities v_t is defined as

$$290 \quad \Psi(\tau) = \sum_t v_t v_{t+\tau} \quad (1)$$

with the temporal shift τ and the vertical velocity v at time t . To compare different cloud ~~objects~~ cases, the autocorrelation function $\Psi(\tau)$ is normalized with $\Psi(0)$. The temporal shift τ from the observations is converted into a horizontal shift or autocorrelation length l with the Cloudnet model-based horizontal wind velocity v_{hor} :

$$\Psi(l) = \Psi(\tau) v_{\text{hor}} \quad (2)$$

295 Similarly the ~~vertical-velocity~~ vertical-velocity spectral power density is calculated by a Fast Fourier Transform of the ~~vertical-velocity~~ vertical-velocity time series. High autocorrelation coefficients for large shifts and low power density are indications for wave-driven, ~~low-turbulent~~ low-turbulent flow. The inset in Fig. 2c shows the autocorrelation function for each of the identified cloud ~~objects~~ cases. All of them are weakly affected by gravity waves, but ~~small-scale~~ small-scale turbulence dominates. ~~When the cloud phase~~ In contrast, the wave cloud in Fig. A1d shows high autocorrelation coefficients for longer
300 shifts. As a characteristic value of the autocorrelation function, the shift at which the coefficient drops below 0.8 was chosen after visually inspecting the whole dataset. When the ice-formation frequency statistics is investigated for an influence of gravity waves (Sec. 3.2.3), the ~~length-scale, where the autocorrelation drops below 0.8 is used to separate wave-driven clouds from clouds with fully developed turbulence~~ shift threshold is reduced step by step. As the autocorrelation function is generally decreasing, a lower characteristic value than 0.8 would result in larger shift thresholds. For shifts larger than about 500 m,
305 random fluctuations appear for clouds with a rapid drop in autocorrelation coefficients. Thus, 0.8 is a robust choice for the characteristic value.

3 Results

In this section, the thermodynamic phase partitioning and quantitative ice mass production in ice forming shallow cloud layers are presented and compared between all measurement sites under study. First, the average profiles of aerosol optical and microphysical properties are presented. Then, instrumental detection thresholds, boundary layer effects and gravity wave activity are all analyzed as potential influencing factors on the retrieved ~~ice-formation~~ice-formation characteristics.

3.1 Aerosol conditions at Leipzig, Limassol, and Punta Arenas

To provide a general insight into the aerosol conditions at the three sites, the average optical and microphysical aerosol properties as derived from the lidar observations (Sec. 2.2) are shown in Fig. ~~3~~3 and Fig. ~~4~~4. The impact of aerosol on clouds is controlled more strongly by temperature than geometrical height, hence the averages are also calculated with temperature as a vertical coordinate.

3.1.1 Optical properties

The average aerosol backscatter coefficient β_p at 532nm and the particle depolarization ratio derived from the Polly^{XT} observations with the Klett method (see Sec. 2.2) are investigated in this section. The evaluation of the air mass sources is covered more quantitatively in the next section and Appendix B.

The central European site of Leipzig is characterized by predominantly continental aerosol mixed with anthropogenic pollution (Baars et al., 2016). Long-range transport of dust may occur periodically, especially during spring and autumn (Ansmann et al., 2003) as well as lofted smoke layers from wildfires (~~Haarig et al., 2018~~)(Haarig et al., 2018; Baars et al., 2021). Aerosol optical thickness (AOT) is derived from sun-photometer observations. Mean AOT at 500nm is 0.198 at TROPOS, Leipzig, ~~between 2018 and 2018 is 0.198, 2014 and 2018~~. When only the periods with co-located Polly^{XT} observations (used in this study) are considered, the AOT is 0.216. Mean β_p at 532nm drops below $0.2 \text{ Mm}^{-1} \text{ sr}^{-1}$ only above 4km height, which corresponds to an extinction coefficient of 1.0 Mm^{-1} ~~for assuming~~assuming a and a corresponding lidar ratio of 50sr.

Limassol is characterized by a distinct dry season with no precipitation and very few clouds during the summer. Generally, Limassol is frequently affected by aerosol transport from Africa, the Middle East and Europe with aerosol characteristics including dust (mineral and soil), marine (organics and sea salt) and anthropogenic pollution as well as mixtures of these (Nisantzi et al., 2015). Mean AOT at 500nm is 0.176 during the whole observational period and 0.165 during the ~~2~~² ‘cloudy season’ from October to May. In the following, the non-cloud season from June to September is excluded from the statistics. The profile of mean backscatter is similar to the one at Leipzig within a factor of 1.5, whereas the median generally is higher at Leipzig.

The aerosol load at Punta Arenas can be separated into two distinct layers with an aerosol-rich boundary layer and pristine conditions aloft. The free troposphere is dominated by marine aerosol from the Southern Ocean and was reported to show no changes compared to the pre-industrial conditions (Hamilton et al., 2014). Nevertheless, events of aerosol long-range transport

also occur occasionally (Foth et al., 2019; Floutsi et al., 2021). The boundary layer is laden with a mixture of marine and
340 continental aerosol, as Punta Arenas is located 230km inland from the Pacific coast. Mean AOT at 500nm is 0.055 during
the whole campaign, but dropping to 0.047, when excluding the period of long-range wildfire smoke transport in early 2020
(Ohneiser et al., 2020). Average boundary layer height is around 1.5km (Foth et al., 2019) with negligible β_p above 2.0km
height (90% percentile of β_p at 532nm dropping below $0.2\text{Mm}^{-1}\text{sr}^{-1}$). Comparing the backscatter at Punta Arenas and
Limassol, the 90% percentile at Punta Arenas is more than 30% below the mean of Limassol and Leipzig at similar heights
345 above ground. Both European sites show quite some variability, as well, with the 90% percentile β_p twice as large as the mean.

At temperatures above 5°C , β_p shows the strongest difference between the three sites (Fig. 3b), also explaining the larger
AOT at Limassol and Leipzig. Between 0°C and -12°C , the mean β_p at Limassol and Punta Arenas are almost equal. This
counterintuitive behavior can be explained by different temperature regimes. The -5°C isotherm at Punta Arenas varies be-
tween 1.1 and 3.3km height, whereas at Limassol it varies between 3.0 and 4.8km height and at Leipzig between 2.1 and
350 4.8km. At Punta Arenas, corresponding to the height of 2.0km, only very low values of β_p are observed at heights below a
temperature of -10°C . The pronounced decrease of backscatter at Leipzig and Limassol is observed at slightly higher tempera-
tures, which agrees with the, on average, higher boundary layer temperature there. Assuming typical lidar ratios of ~~50, 45, 20sr~~
50 (continental), 45 (dust), and 20sr (marine) at Leipzig, Limassol, and Punta Arenas, respectively, typical aerosol extinction
coefficients can be estimated from the median β_p profile (Sec. 2.2). As shown in Fig. 3c, the extinction coefficients at Punta
355 Arenas are a factor 2-3 lower than over Limassol for temperatures below -10°C . This difference decreases to a factor of
1.5 for slightly supercooled ~~temperatures-conditions~~ of above -10°C . Comparing Punta Arenas and Leipzig, the extinction
is a factor of 3-4 higher at the latter site for all temperatures below freezing. This optical properties serve as a proxy for the
background reservoir of aerosol particles that could act as cloud condensation nuclei and ice nucleating particles in the free
troposphere.

360 The particle depolarization ratio provides hints on the aerosol particle types (Fig. 3d). At Leipzig, the depolarization ratio
is approximately 0.05 at all temperatures, typical for a continental aerosol with a slight contribution of mineral dust. The
depolarization ratio at Limassol is bimodal, with one peak above 20°C , a second one at -14°C and a minimum at 4°C . The
first ~~one-peak~~ can be ascribed to mineral dust in the boundary layer and originating from local sources. The second one at
is caused by long-range transport of mineral dust. At Punta Arenas, a maximum at $+8^\circ\text{C}$ is caused by occasional events of
365 dried sea salt aerosol at the top of the atmospheric boundary layer (Haarig et al., 2017; Bohlmann et al., 2018). Going to lower
temperatures, the depolarization ratio has a minimum of 0.01 at -20°C and a slight increase afterwards.

3.1.2 Contrasts in INP concentrations

An important aspect for the discussion of microphysical contrasts in clouds over the three sites is ~~;~~ how the differences in
aerosol optical profiles are linked to contrasts in the INP load. An estimate of average INP concentrations covering the whole
370 troposphere can be derived with the parametrizations described in Sec. 2.2. The decision for the aerosol typing required in the
INP retrieval is based on the particle depolarization ratio and air mass source estimates. The air mass source methodology is
based on Radenz et al. (2021) and shown in Appendix B.

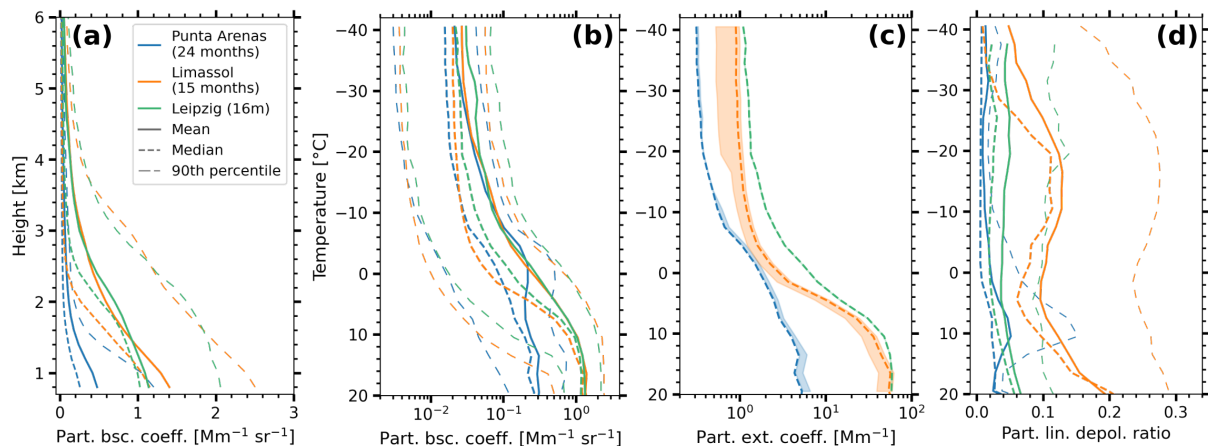


Figure 3. Profiles of average aerosol optical properties at 532nm wavelength derived by the PollyNET retrieval using the Klett method. **Partiele**-(a) **particle** backscatter coefficient over height (a)-and **temperature**-(b) **temperature** for Leipzig, Limassol, and Punta Arenas. **Extinction coefficient**-(c) **Extinction coefficient** and **partiele depolarization ratio**-(d) **particle depolarization ratio** for the same locations. For the extinction coefficient only the median values based on the typical lidar ratio at each site are shown. Variability caused by different aerosol mixtures (see Sec. 3.1.2) is denoted by shading.

At Punta Arenas marine sources are by far the most frequent, contributing 90% to the residence time throughout the troposphere and peaking 95% at 2.7km height. Only below 2.0km height (or above -10°C) local terrestrial sources make up to 10% of the air mass. Above 5km height (below -20°C) sparsely vegetated areas in Australia contribute up to 4% to the air mass source. Mean particle depolarization ratios below 0.02 in the free troposphere exclude frequent presence of mineral dust (Fig. 3d). Contrarily, at Limassol air masses with a marine source contribute 50-50% to 80% to the mixture. But these air masses are not pristine marine, as the eastern Mediterranean is enclosed by strongly populated landmasses. Below 5km height (above -16°C) continental Europe is second strongest source, responsible for up to 45% at 0.5km height. In the upper troposphere, barren ground from the Sahara is the strongest terrestrial source with contributions around 15%. POLIPHON (see Sec. 2.2) shows a peak at -12°C with mean dust fractions of 0.3 (90% percentile 0.87). Hence, the backscatter is divided into dust and non-dust according to the dust fraction. The non-dust portion is then split up into continental and marine, with the 40% contribution of the continent above -12°C and 20% below. The aerosol mixture at Leipzig is dominated by continental aerosol, with an average dust fraction of 0.1.

The derived INP concentrations are depicted in Fig. 4. Note, ~~shown is that~~ the INP concentration at a certain atmospheric temperature (~~also as~~ a proxy for height) ~~is shown~~ based on the optical properties, aerosol types and the parametrization, and the parameterization, not a freezing spectrum obtained from sampling a single air parcel and varying temperature.

At temperatures above -10°C average INP concentrations between ~~$4 \cdot 10^{-3}$ to $6 \cdot 10^{-2} \text{L}^{-1}$~~ 4×10^{-3} to $6 \times 10^{-2} \text{L}^{-1}$ can be expected at all three locations. With decreasing temperatures, the concentration increases to $0.1 - 1 \text{L}^{-1}$ at -25°C at the northern-hemispheric sites of Leipzig and Limassol. A strong increase of ice nucleating efficiency with decreasing temperature

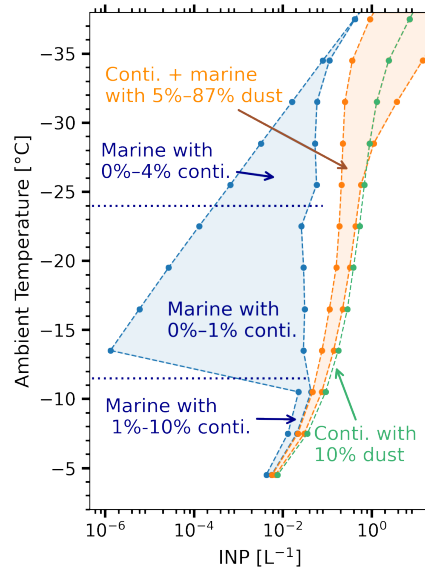


Figure 4. Average INP concentrations derived from optical properties for Leipzig (green), Limassol (orange), and Punta Arenas (blue). Shading shows the spread covered by different aerosol compositions, similar to Fig. 3c. Temperature is used as a vertical coordinate. The fraction of continental aerosol is abbreviated ‘conti.’. The used parametrizations are described in Sec. 2.2.

is counterbalanced by a decreasing aerosol concentration at lower temperatures, which are tied to greater heights. INP concentrations at Punta Arenas are strongly controlled by the fraction of continental aerosol in the free troposphere. Continental sources can contribute up to 1% between -11 and -24°C and up to 4% at lower temperatures. When only pristine marine aerosol is present, the INP concentration can be expected to be 3-4 orders of magnitude lower than at the two other sites. Equally low concentrations of INP were also found by McFarquhar et al. (2021) in the remote Southern Ocean south of Australia. As soon as very small fractions of continental aerosol are present, the INP concentration increases significantly. However, even if few percent of continental aerosol are assumed, throughout the heterogeneous freezing regime, INP concentrations remain a factor of 2-6 lower at Punta Arenas, compared to Leipzig and Limassol. Due to the absence of suitable remote-sensing or in-situ measurements, the actual contribution of continental aerosol to the free-tropospheric aerosol load over Punta Arenas can to date not be obtained. The range given in Fig. 4 is thus a solid estimate of the minimum-and-maximum-of-the expected range of possible INP values concentrations. At temperatures above -10°C , which refers to a height range that is frequently within the boundary layer, INP concentrations at Punta Arenas are high and within an order of magnitude to the concentrations at Leipzig and Limassol at the same temperature. Concentrations that are equally high as retrieved from the remote sensing observations above -10°C were also found in-situ-at-in-situ at the up-wind hilltop station on Cerro Mirador in an altitude of 622 m above sea level (Gong et al., in preparation).

3.2 Mixed-phase stratiform cloud properties

The automated stratiform cloud selection algorithm introduced in Sec. 2.4 is applied to the Cloudnet datasets of all three locations. An overview over identified clouds and temperature-resolved phase occurrence frequency is provided in Sec. 3.2.1. Afterwards, it is shown how boundary layer aerosol load (Sec. 3.2.2) and gravity waves affect the derived-phase occurrence
 410 (Sec. 3.2.3). In Sec. 3.2.4 contrasts of ice content in the virga are briefly discussed.

3.2.1 Phase occurrence frequency and detection thresholds

Fig. 5 shows all the cloud objects-cases that fulfill the selection criteria introduced in section 2.4. 2.4. The time share of ice virga sedimenting out of the liquid-dominated top is given by the fraction of profiles for which ice was classified below the liquid layer. Values range from 0 (no ice virga observed at all) to 1 (ice virga observed all the time). At all three locations, the fraction
 415 of profiles (i.e., fraction of time) in which a virga originated from the liquid layer increases with decreasing temperature. Similarly, the ice-to-liquid content ratio (Bühl et al., 2016) increases for lower temperatures. Liquid only clouds (fraction of profiles where no ice was produced from the liquid layer) were observed at Leipzig and Limassol down to -16°C . Whereas at Punta Arenas such clouds were observed even at temperatures as low as -38°C .

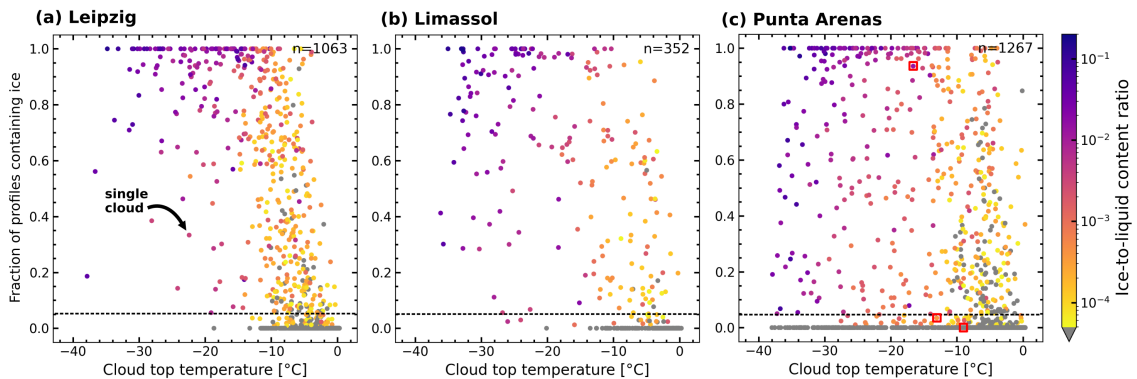


Figure 5. All stratiform clouds identified by the automated selection algorithm for Leipzig (a) Leipzig, Limassol (b) Limassol, and Punta Arenas (c) Punta Arenas by their cloud top temperature and the fraction of profiles, where ice is observed below the liquid-dominated cloud top. The 0.05 threshold for the classification is marked by a dashed line. The color gives the median ice-to-liquid content ratio. n gives the number of cloud objects-cases in each dataset. Red rectangles-squares in (c) denote the clouds also shown in Fig. 2.

The First, the temperature-resolved fraction of occurrence of ice-forming clouds provide a first insight into the contrasts
 420 between different locations (Choi et al., 2010; Kanitz et al., 2011; Seifert et al., 2015; Tan et al., 2014; Zhang et al., 2018) is depicted in Fig. 6. For the phase occurrence frequency, a cloud is classified as ice-producing, if an ice virga was observed at least during 5% of a cloud objects duration the duration of a cloud case (Sec. 2.4).

Generally, more Comparing the frequency of ice-containing clouds at different locations provides insights into differences of primary ice formation (Choi et al., 2010; Kanitz et al., 2011; Seifert et al., 2015; Tan et al., 2014; Zhang et al., 2018). Generally,

clouds contain ice more frequently for decreasing temperature (Fig. 6, solid lines). While at Leipzig and Limassol nearly all clouds with CTTs below -16°C contained ice, at Punta Arenas a fraction of $0.5-0.7$ 0.4 ± 0.1 of shallow stratiform clouds at these temperatures were classified as liquid only. This behavior is discussed further in Sec. 3.2.3.

Compared to prior lidar-based studies (e.g. Kanitz et al., 2011; Choi et al., 2010; Seifert et al., 2010) (e.g., Kanitz et al., 2011; Choi et al., 2010), the fraction of ~~ice-containing~~ ice-containing clouds in the synergistic dataset is higher at temperatures above -10°C . At these temperatures, the amounts of ice produced and hence, the radar reflectivity and optical extinction are usually very low and stays undetected for lidar (Bühl et al., 2013a) and space-borne radars (Bühl et al., 2016). ~~By utilizing established relationships between, radar reflectivity factor, temperature, IWC and~~ To quantify a lidar detection threshold in terms of optical extinction, ~~such as the one from Hogan et al. (2006), one can estimate the sensitivity of a deployed lidar system to detect a certain mass of ice.~~ In the case of the reflectivity-to-IWC and the reflectivity-to-extinction relationships by Hogan et al. (2006) are used.

The only additional information needed for both parametrizations is the ambient temperature. Using these relationships, the response of the occurrence statistics to arbitrary extinction detection thresholds can be tested. As shown in Fig. 6 (dashed lines), for the lidar data used in the Cloudnet classification, an extinction threshold of 12Mm^{-1} had to be applied in order to best match the lidar-only statistics from Punta Arenas and Leipzig presented by Kanitz et al. (2011) (Fig. 6, dashed lines).

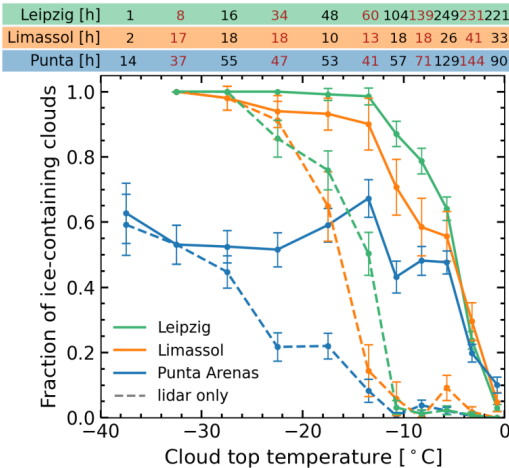


Figure 6. Fraction of ice containing clouds over temperature for Leipzig, Limassol, and Punta Arenas. Total duration of the clouds in each bin is given by the numbers on top in hours. Dashed curves mark the occurrence frequency when using a lidar detection threshold of 12Mm^{-1} .

3.2.2 Effect of boundary layer aerosol load on phase occurrence

As discussed in Sec. 3.1, the aerosol load at Punta Arenas is confined to the lowermost 2 km and the aerosol load at temperatures above -10°C is similar to Limassol. To check for possible impact of this boundary layer aerosol on the ice-formation efficiency, the basic temperature-resolved phase occurrence frequency (Fig. 6) is split into two subsets, one containing cloud objects-cases with bases below 2 km height and one with cloud objects-cases having bases above that thresh-

old(Fig. 7), in the following denoted as coupled and uncoupled clouds, respectively. The resulting ice-formation frequency is shown in Fig. 7. At any height, temperatures vary by more than 11°C (~~10-10%~~ to 90% percentile), providing ample coverage for the height threshold. Generally, coupled clouds show higher fractions of ice (absolute difference increases by 0.3 at all locations at -8°C). The temperature-resolved phase occurrence frequency for the coupled clouds show rapid increase in fraction of ice-containing clouds, reaching 1.0 at temperatures of only -15°C . The absolute difference between the fractions is less than 0.20, with the lowest fractions still being observed at Punta Arenas. Below -15°C , almost no clouds were observed at such low heights, especially at Limassol, hence no meaningful comparison can be done in this temperature interval.

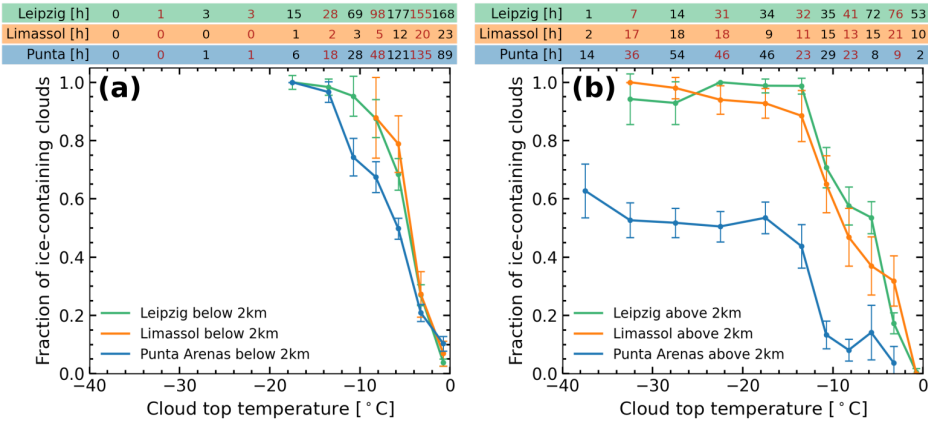


Figure 7. Fraction of ice containing clouds over temperature for Leipzig, Limassol, and Punta Arenas, (a) below 2km and (b) above 2km. Total duration of the clouds in each bin is given by the numbers on top in hours.

Considering only uncoupled clouds (cloud base above 2km height, Fig. 7b), stronger contrasts become evident. The fractions wave clouds discussed in the following section are still included and impact the frequency at temperatures below -18°C at Punta Arenas. The frequencies of ice-forming clouds are similar at Leipzig and Limassol, but 0.15 to 0.5 at -10°C lower at Punta Arenas. Comparing the coupled and uncoupled state at Leipzig and Limassol, ice formation is more frequent in the coupled case by 0.15 between -5 and -12°C .

3.2.3 Gravity wave influence on phase occurrence at low temperatures

The lack-lower frequency of ice-containing cloud layers at temperatures below -18°C over Punta Arenas is a prominent feature of both, the lidar-radar and the lidar-only-equivalent datasets shown in Fig. 6. The associated excess-high abundance of supercooled liquid water is frequently reported as a general phenomenon of stratiform clouds in the southern hemisphere mid and higher latitudes. Within this subsection, the reasons for the found behavior over Punta Arenas will be elaborated in more detail.

Frequently, the stratiform liquid-only cloud layers observed over Punta Arenas at temperatures below -18°C are embedded in orographic gravity waves. Following the wave detection methodology introduced in Sec. 2.5, Fig. 8 shows the autocorrelation

and power spectra of the Doppler lidar vertical velocity for clouds classified as liquid only over Leipzig, Limassol, and Punta Arenas, respectively. These supercooled liquid-only clouds at Punta Arenas show high autocorrelation coefficients at long shifts (Fig. 8c1), whereas liquid only clouds with similar characteristics are absent at Limassol (Fig. 8b1) and Leipzig (Fig. 8a1). In terms of spectral power density (Fig. 8c2), the strongly supercooled clouds at Punta Arenas show only low turbulence.

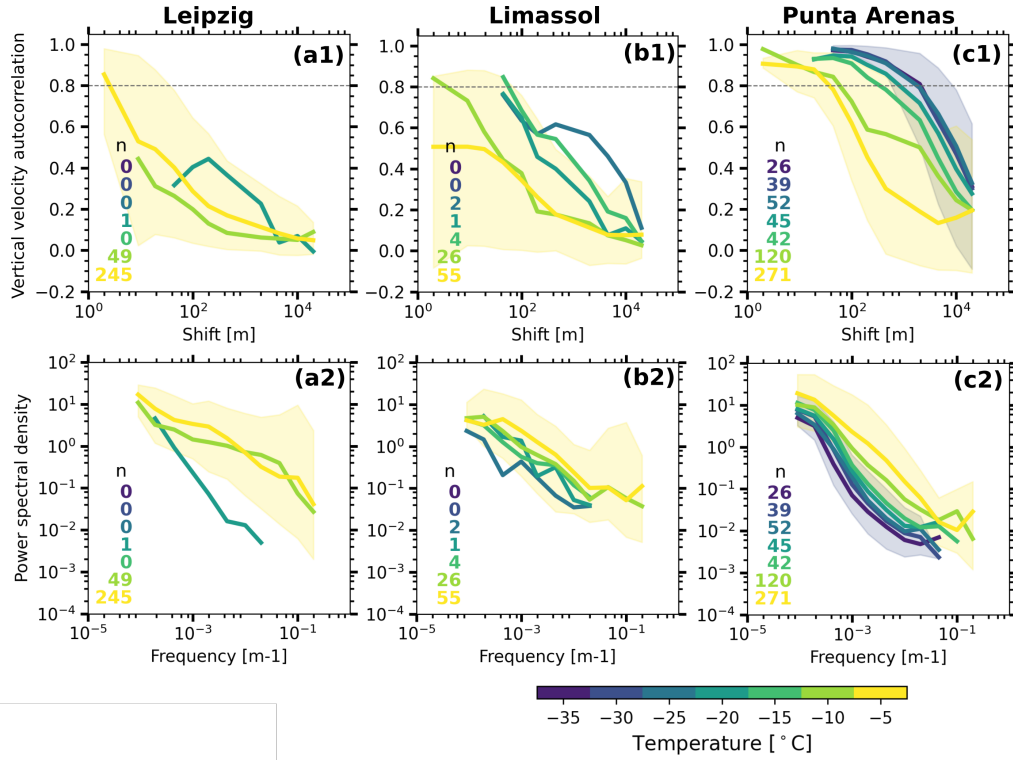


Figure 8. Autocorrelation-Vertical-velocity autocorrelation function (a1, b1, and c1) and power density (a2, b2, and c2) of the Doppler lidar vertical velocities for cloud objects-cases classified as liquid-only liquid-only. The columns are Leipzig (a) Leipzig, Limassol (b) Limassol, and Punta Arenas (c) Punta Arenas. Color gives cloud top. The curves are binned to CTT intervals of 5 K with color indicating the CTT. The number of clouds for each temperature interval is given as numbers in the respective panel. Shading denotes the 10%-90% percentile range for the -5 and -30 °C bin.

By decreasing the length threshold In a next step, the autocorrelation is used to filter the dataset for clouds affected by gravity waves. As described in Sec. 2.5, the length at which the autocorrelation drops below 0.8, weakly wave-influenced clouds are gradually removed from the phase-occurrence-frequency-statistics coefficient drops below 0.8 is used as a characteristic value. Decreasing threshold value for this characteristic length will remove gravity-wave influenced clouds from the dataset. For large values of the length threshold, e.g., larger 1000 m, only clouds that are strongly forced by gravity waves will be removed, whereas going to shorter thresholds (< 500 m) will also remove weakly gravity-wave-influenced clouds. Fig. 9 shows an increase in the fraction of ice-containing clouds below -12 °C with decreasing correlation-length thresholds from 30000

475 correlation-length thresholds from 30000 to 300 m. At Punta Arenas, the fraction ~~increases from 0.5 to 0.85~~of ice-containing clouds increases from 0.5 to 0.85. In the temperature interval between -15 and -25°C ~~-12°C~~ , the fraction is ~~0.05 to 0.1~~ 0.05 to 0.1 lower at Punta Arenas compared to Leipzig and Limassol. Hence, clouds with fully developed turbulence show ~~similar ice-formation frequencies, independently~~ similar ice-formation frequencies, independent of the location, with indications for a still slightly reduced ~~ice-formation~~ ice-formation efficiency over Punta Arenas.

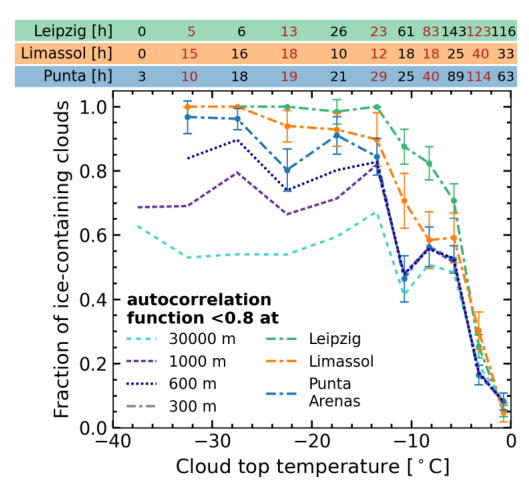


Figure 9. Fraction of ice containing clouds over temperature for Leipzig, Limassol, and Punta Arenas. Dash-dotted lines show the fractions for Leipzig, Limassol, and Punta Arenas with an autocorrelation coefficient smaller than 0.8 for a horizontal shift of 300m. The fractions for cloud ~~objects-cases~~ with longer autocorrelation are only shown for Punta Arenas. Total duration of the clouds in each bin is given by the numbers on top in hours.

480 **3.2.4 Comparison of radar reflectivity factor of the ice virga**

Prior studies of Zhang et al. (2018) identified a strong contrast in radar reflectivity factor between the different 30-deg latitude bands of the globe, with the southern hemisphere mid-latitudes (30-60°S) showing the lowest mean reflectivity of all regions. They concluded that this difference in reflectivity factor is associated to a respective difference in ice crystal mass and number concentration. In the following we provide a similar representation of regional contrasts of ice-virga reflectivity from ground-
485 based perspective that is based on a single radar instrument. As described in Sec. 2.4, the amount of ice formed in the mixed phase layer is measured at six height bins (180m) below the base of the liquid dominated cloud top and hence at the top of the virga (Bühl et al., 2016). Fig. 10a shows the cloud top temperature-resolved statistics of reflectivity for the three stations, which is based on the full cloud dataset, including the wave-influenced clouds ~~(as these are also included in the study by Zhang et al. (2018))~~ (as these are also included in the study by Zhang et al., 2018). From -28°C to -16°C and again above
490 -12°C , Punta Arenas shows the lowest reflectivity and Limassol the highest. For most temperatures, Punta Arenas is 5 – 8 dB below the northern hemispheric stations, only exception is the interval between -4 and -8°C , where the reflectivity for Punta

Arenas and Leipzig is almost equal. At temperatures below -28°C , Limassol and Punta Arenas show equal reflectivity with values at Leipzig being slightly higher.

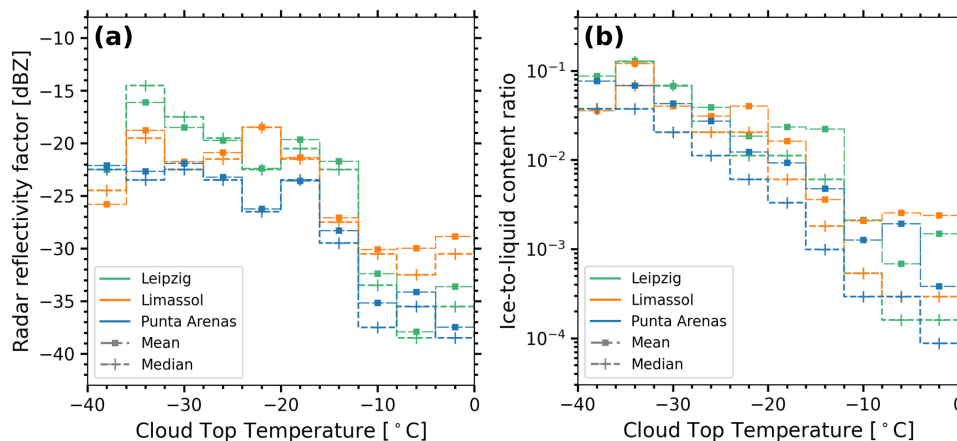


Figure 10. Radar Amount of ice observed in the virga, (a) radar reflectivity factor observed at the top of the virga (180 m below the liquid-dominated layer base) (a) and ice-to-liquid content ratio (b) ice-to-liquid content ratio binned by CTT. Temperature bins of 4°C .

We further investigated the properties of the ice-forming liquid-dominated cloud top layers and found that cloud thickness agrees within 40 m above -30°C . The ice-to-liquid content ratio (Fig. 10b) is smaller at Punta Arenas, than at the northern hemispheric locations, especially (factor 3) between -24 and -20°C , but also above -10°C . Hence, in these temperature regimes, the liquid phase is less efficiently converted into solid ice in stratiform clouds above Punta Arenas.

4 Discussion of observed contrasts in properties of shallow clouds

In the previous section, a comprehensive overview about of the aerosol conditions and stratiform cloud properties at the strongly contrasting sites of Leipzig, Limassol, and Punta Arenas was presented. Several aerosol-, temperature-, surface-surface-coupling-, as well as dynamics-related differences have been identified and will be discussed in the following.

The analysis of the profiles of aerosol optical properties obtained from the Polly^{XT} lidar observations revealed-reveals almost similar aerosol load at Punta Arenas and Limassol between 0 and -10°C . Leipzig shows a higher aerosol load in this temperature interval. For lower temperatures the lowest average extinction was observed at Punta Arenas. Low values of particle depolarization ratio reveal-indicate that non-spherical particles, such as mineral dust are completely absent at Punta Arenas below -10°C . The absence of continental aerosol species in the free troposphere is also consistent with the Southern Ocean south of Australia (e.g., Alexander and Protat, 2019) (e.g., Alexander and Protat, 2019). Additionally, Uetake et al. (2020) showed for this region that INP are related to sea spray emissions. Together with an air mass source estimate, the average profiles of optical parameters were used as an input for INP parametrizations for a first estimate of long-term averages of INP concentrations. The lidar-based estimate of average INP concentrations identifies the strongest differences in INP concentrations between -12

and -35°C . However, it is still subject to discussion which fraction of marine particles is transported from the boundary layer to the free troposphere. This insufficient knowledge propagates further into uncertainties in the fraction of continental aerosol, which leads to considerable uncertainties in the estimated concentrations of INP. Bourgeois et al. (2018) argument ~~that~~ due to efficient removal processes, marine particles predominantly remain in the boundary layer (globally 80% of their AOT). Murphy et al. (2019) ~~argument~~ argue in a similar direction, especially when sea salt is regarded a tracer of marine origin.

Opposed to prior lidar-only studies (~~e.g. Kanitz et al., 2011; Alexander and Protat, 2018~~) (e.g., Kanitz et al., 2011; Alexander and Protat, 2018), more than half of the clouds observed at -8°C at the three sites were found to contain ice. This is in line with the lidar detection threshold introduced by Bühl et al. (2013a), but extending their results to Limassol and Punta Arenas. This result has significance for lidar-only or ceilometer datasets that are currently used for validation of climate models (~~e.g. Kuma et al., 2020~~) (e.g., Kuma et al., 2020). The occurrence of ice at these relatively warm temperatures is also in line with airborne in-situ observations over the Southern Ocean (Huang et al., 2017; D'Alessandro et al., 2019).

Ice formation at slightly supercooled temperatures-conditions above -10°C was found to be equally frequent, suggesting the presence of equally efficient INP at all sites. At Punta Arenas, such temperatures usually occur within the boundary layer, where presumably INP of biological origin are present (Gong et al., in preparation). Similarly high fractions were also observed for Arctic boundary layer clouds during summer and autumn (Achtert et al., 2020). Griesche et al. (2020) also found ~~that~~ surface coupling increased the frequency of ice formation in boundary layer clouds observed during Arctic summer.

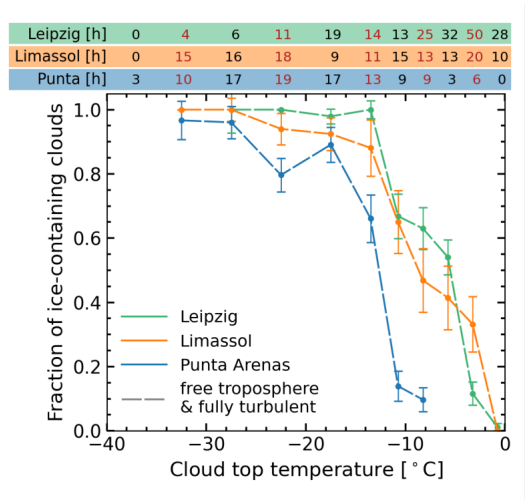


Figure 11. Fraction of ice containing clouds over temperature for Leipzig, Limassol, and Punta Arenas when considering only fully turbulent clouds with an autocorrelation coefficient smaller than 0.8 for a horizontal shift of 300 m (see Fig. 7) and cloud bases in the free troposphere above 2 km height (see Fig. 9).

The liquid-only clouds at Punta Arenas found below -14°C are identified to be predominantly mountain-wave-clouds-wave clouds forced by orography, likely in an early period of their lifecycle. In this kind of cloud, the liquid-dominated mixed-phase is separated horizontally from the ice virga, due to the rapid flow and weak turbulence (Heymsfield and Miloshevich, 1993).

530 When sampled by stationary observations, only parts of the clouds are covered and the phase occurrence frequency is biased. Using Doppler lidar vertical velocity observations, clouds driven by orographic wave dynamics can be identified based on their autocorrelation function, as presented in Sec. 3.2.3. At temperatures below -14°C , approximately ~~1/3~~one-third of the cloud layers observed at Punta Arenas were influenced by gravity waves. Similar frequencies of occurrence of non-turbulent liquid layers were also found at Utqiagvik (Alaska) and McMurdo (Antarctica) deriving stability criteria from radiosoundings (Silber
535 et al., 2020).

In a final step, the separation techniques for coupling and orographic waves can be combined, to assess contrasts ice frequency for free-tropospheric and ~~fully-turbulent~~fully-turbulent clouds. The resulting occurrence frequency is shown in Fig. 11. The fraction of ~~ice-containing~~ice-containing clouds at temperatures below -15°C is above 0.85 at all three sites. However, comparing to Leipzig and Limassol, the fraction of ice forming clouds with CTT between -25 and -15°C remains 0.1 lower
540 at Punta Arenas. Mineral dust is known to be an efficient INP at these temperatures (Kanji et al., 2017), but was not observed at the respective temperatures at Punta Arenas (Fig. 3). This difference in the non-wave cloud phase occurrence is also in agreement with satellite-based studies of Villanueva et al. (2020), who also attributed latitudinal differences in the ice occurrence to associated differences in the dust load. Fig. 11 also depicts the low frequency of ice formation in free-tropospheric ~~fully~~
~~turbulent~~fully-turbulent clouds at temperatures above -15°C . With less coupling to the near-surface aerosol reservoir, ice
545 formation is strongly suppressed compared to Leipzig and Limassol.

The ice mass formed by stratiform liquid layers is lowest at Punta Arenas, as was found by investigating the radar reflectivity factor in the ice virga as a proxy. Especially between -28°C and -16°C , the observed radar reflectivity factor is up to 7 dB lower, compared to both sites in the northern hemisphere. This result is consistent with estimates from space-borne sensors covering the full Southern Ocean (Zhang et al., 2018). Contradicting, Arctic mixed-phase clouds were found to respond with
550 lower IWC to increased loads of anthropogenic pollution (Norgren et al., 2018). As for the lower frequency of ice formation discussed above, the difference in ice mass coincides with a lack of dust INPs at these temperatures. Slight differences in IWC or Z, respectively, might be explained by a slower mass growth rate, caused by a smaller vapor diffusion coefficient at higher ambient pressure (Hall and Pruppacher, 1976). When temperature and particle size are considered similar, the stratiform clouds subject to this study, experience ~~10-10%~~10-10% to 20% larger growth rates at Leipzig and Limassol than at Punta Arenas. Hence, the
555 difference of a factor 3-6 larger ice mass cannot be explained solely by this effect alone.

Frequent occurrences of ice forming clouds above -10°C were found, which were not covered by studies based on space-borne active remote sensing. Similar to their colder counterparts, they show a lower ice amount in the virga above Punta Arenas, but with smaller difference compared to the sites in the northern hemisphere. With an average reflectivity of -36 dBZ, they are usually well below the detection limit of the ~~CLOUDSAT~~CloudSat satellite in the A-Train constellation (Bühl et al.,
560 2013a, 2016). Occurrence of such clouds in other parts of the Southern Ocean cannot be ruled out. A misclassification of super-cooled drizzle clouds as ice containing is unlikely, as they exceed -30 dBZ neither at cloud top, nor in the virga. From previous studies it is known that the onset of drizzle formation is usually associated to higher reflectivities, either above approximately -20 dBZ (Liu et al., 2008; Acquistapace et al., 2019) or at least above -30 dBZ (Wu et al., 2020).

5 Summary and Outlook

565 This study investigated contrasts in aerosol-cloud interactions in shallow supercooled stratiform clouds observed with the ground-based remote sensing supersite LACROS at Leipzig, Limassol, and Punta Arenas.

Sampling the profiles of optical properties with temperature as a vertical coordinate revealed aerosol load at temperatures between -15 and 0°C being within a factor of 2 at Punta Arenas and Limassol. This finding is related to the cold and (compared to the free troposphere) aerosol laden boundary layer at Punta Arenas. At lower temperatures, the lowest β_p and extinction was
570 observed over Punta Arenas, the highest over Leipzig. The very low particle depolarization ratio at Punta Arenas between -25 and -10°C , suggests the absence of mineral dust in a temperature regime, where dust is known to be an efficient INP. An estimate of INP concentrations at the respective temperatures based on the optical properties reveals differences of 1-4 orders of magnitude between Punta Arenas and the two northern hemispheric sites. In absence of abundant INPs from marine sources, the contribution of terrestrial sources causes strong variability.

575 The phase occurrence frequencies showed a higher fraction of ice containing clouds at weakly-supercooling temperatures of above -10°C compared to prior lidar-only studies. A cloud radar with a sensitivity better than -40dBZ is needed to sufficiently characterize low ice water contents in the virga formed by shallow stratiform clouds in this temperature regime. Coupling to the boundary layer increases the frequency of ice formation at slightly ~~supercooled~~-supercooling temperatures at all sites. The strongest contrasts in the ~~ice-formation~~-ice-formation frequency between free-tropospheric and surface-coupled
580 conditions were found for Punta Arenas. This finding is in compliance to the found contrasts in the INP profiles at the three sites and further indicates that the free-tropospheric INP reservoir over the Southern Ocean is limited.

Frequent liquid only layers below -20°C at Punta Arenas were found to be associated with orographic gravity waves, causing two implications: (1) potential phase misclassification by stationary observers due to horizontal separation of ice and liquid phase; (2) sustained liquid water in updrafts, because the associated vertical velocities allow supersaturation over water;
585 The newly developed Doppler lidar autocorrelation approach helps to address "[...] the difficulty in characterizing gravity waves from existing single-site measurements [...]" as posed by Silber et al. (2020). Ice mass in the virga and the ice-to-liquid content ratio was found to be lowest at Punta Arenas, especially at temperatures where dust serves as an efficient INP at the northern hemispheric sites.

From the results of this study, two issues for further investigations arise. Firstly, importance of terrestrial sources of INP
590 in the Southern Ocean. Observations of free-tropospheric aerosols upwind and downwind of major landmasses are required, to quantify the terrestrial emission and identify regions, where the emitted INP impact cloud microphysics the strongest. Secondly, how important are gravity waves in the formation of clouds in other regions of the Southern Ocean? How frequent do gravity waves occur above the open ocean, which covers the vast majority of the Earth's surface between 30 and 70°S ? Such measurements are required in order to further constrain the role of INP in the evident excess of liquid water in clouds of the
595 ~~southern hemisphere~~-southern-hemispheric mid-latitudes.

Code and data availability. The Cloudnet datasets are provided by the ACTRIS Data Centre node for cloud profiling under the following links: <https://hdl.handle.net/21.12132/2.e130b236928a4932> (Leipzig), <https://hdl.handle.net/21.12132/2.a056a828a6b94d1f> (Limassol) and <https://hdl.handle.net/21.12132/2.b6c194d7d33b448e> (Punta Arenas). In the upcoming future, the Doppler lidar and PollyNET datasets will also be available via ACTRIS. Meanwhile, they can be obtained upon request at polly@tropos.de. The source code of the cloud sniffer, the database of identified clouds and analysis scripts are available at https://github.com/martin-rdz/larda_cloud_sniffer (last accessed 27.04.2021; Radenz and Bühl, 2021) and requires a setup of pyLARDA (<https://github.com/lacros-tropos/larda>, last accessed 27.04.2021; Bühl et al., 2021). AERONET photometer observations at Leipzig, Limassol and Punta Arenas are available from the AERONET database (<http://aeronet.gsfc.nasa.gov/>). We thank AERONET-Europe for providing the calibration service.

Appendix A: ~~Air mass source estimate~~Wave-cloud example

Fig. A1 shows a typical wave cloud at Punta Arenas. Updrafts in stationary or slowly propagating waves can sustain liquid layers without showing signatures of sedimenting ice crystals to the fixed observer. The ice phase is only visible downwind as soon as the droplets have evaporated. Such a wave cloud was observed at Punta Arenas on the 27 September 2019. A mid-tropospheric long-wave ridge caused a strong, zonal westerly flow at Punta Arenas. Velocity of the horizontal wind exceeded 25ms^{-1} at 4000m height. Layered clouds developed within the orographic gravity waves triggered by this flow. One of the cloud layers with a CTT between -34 and -31°C is depicted in Fig. A1. It illustrates how liquid and ice phase are horizontally separated. Embedded in a slowly propagating gravity wave, the cloud was advected over the site and the full life cycle could be observed. Based on the Doppler lidar vertical velocity (Fig. A1d), the updraft was present until 05:32 UTC. Afterwards, downward motion with increasing velocity set in. Only after the liquid droplets evaporated at 06:17 UTC, the ice particles can be clearly identified in the remote-sensing observations (Fig. A1b, c).

Appendix B: ~~Air mass source estimate~~

The automated time–height-resolved air mass source attribution described by Radenz et al. (2021) is used to characterize air mass origin for the LACROS observations. As in the original publication, 10-day HYSPLIT ensemble backward trajectories are calculated in intervals of 3 hours and 500m throughout the period of the deployment. For the calculation of the residence times, a reception height of 2km is used. Additionally to the MODIS based ~~land-surface~~ cover classification and the the named geography (individually for Leipzig, Limassol, and Punta Arenas), 30° -wide bands of latitude are used to characterize meridional transport. The assignment of the ~~land-surface~~ cover classed and named geography are depicted in Fig. 2 and 3 of Radenz et al. (2021). The average residence time for each set of surface types is shown in Fig. B1. However, as for the aerosol optical properties, geometrical height is less insightful as temperature. Thus, the residence time at each 3h profile is binned by ambient temperature, using analysis profiles. Then, the average residence time is calculated per temperature bin (Fig. B2).

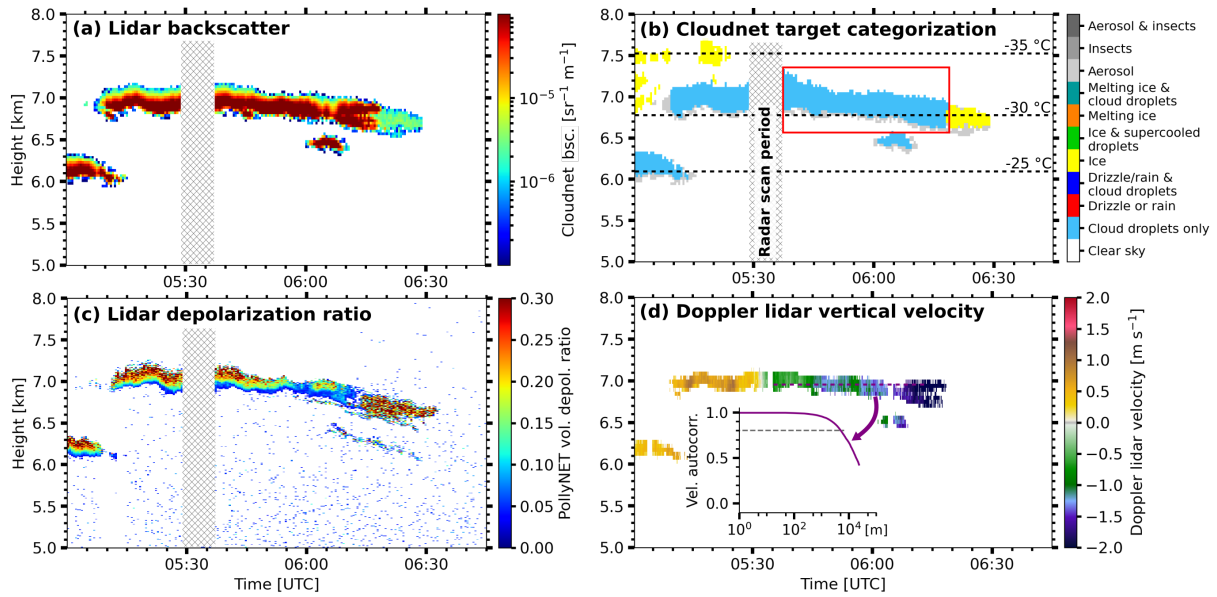


Figure A1. Wave cloud observed at Punta Arenas on 27 September 2019, 05:00–06:45 UTC. (a) Lidar attenuated backscatter, (b) Cloudnet target categorization, (c) Lidar volume linear depolarization ratio and (d) Doppler lidar vertical velocity. The temperature information in (b) is obtained from ECMWF IFS model data. The red rectangle in (d) indicates the automatically detected liquid-topped shallow stratiform cloud. The inset in (d) shows the autocorrelation function of the vertical velocities in the liquid-dominated layer. The height of the velocity sampling is indicated with a dashed line.

625 *Author contributions.* MR analyzed the data and drafted the manuscript. MR, PS, JB, HB, RE, BBG, REM, and FZ conducted the campaigns and operated LACROS. PS, MR, JB generated the Cloudnet dataset. HB processed the Polly^{XT} lidar data. PS, JB, and AA supervised the work and revised the manuscript. All authors jointly contributed to the paper and the scientific discussion.

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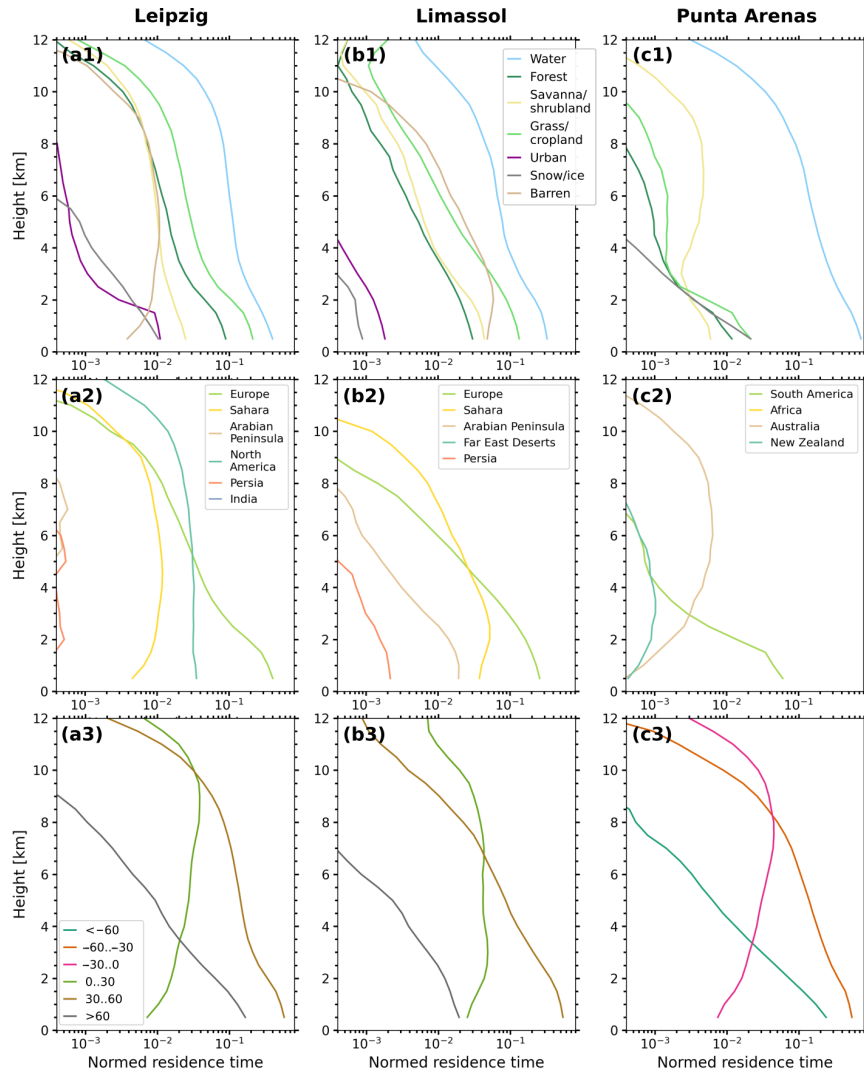


Figure B1. Mean normalized residence time profile at Leipzig (a) Leipzig, Limassol (b) Limassol, and Punta Arenas (c) Punta Arenas for a reception height of 2 km. Residence times are shown for the land surface cover classification (1), named areas (2) and latitude bands (3).

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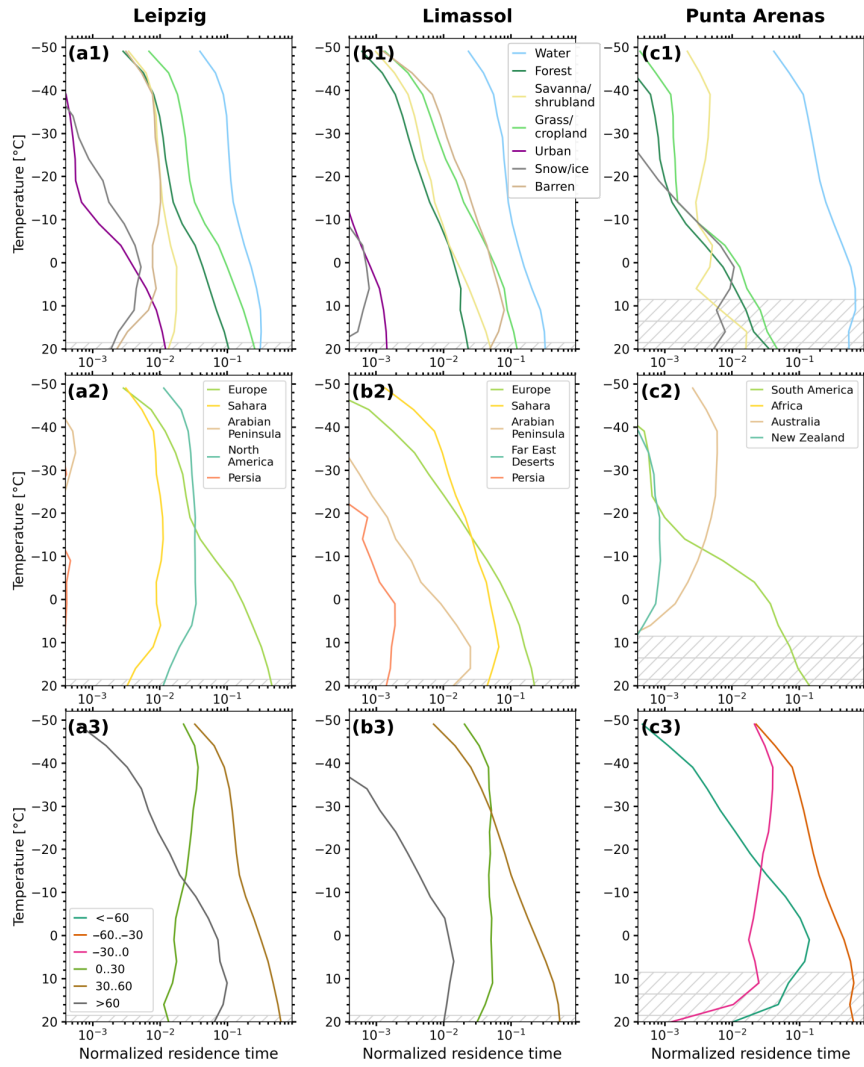


Figure B2. Mean normalized residence time binned by temperature at Leipzig-(a) Leipzig, Limassol-(b) Limassol, and Punta Arenas-(c) Punta Arenas for a reception height of 2km. Residence times are shown for the land surface cover classification (1), named areas (2) and latitude bands (3). Hatching indicates bins with less than 1 % of the values.

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