

## Letter to the Editor

**ACP Discussions doi: 10.5194/acp-2020-552-RC1, ‘Strong variability of the Asian Tropopause Aerosol Layer (ATAL) in August 2016 at the Himalayan foothills’ by Sreeharsha Hanumanthu et al.**

Dear Gabi,

many thanks for handling our manuscript. We prepared and submitted a revised version of our manuscript and are confident that we have satisfactorily addressed all comments of referee s#1 to #2. A detailed point-by-point response to all referee comments (#1 to #2) is attached. Further, a document specifying all changes in the revised manuscript compared to the ACPD version is added.

In addition to the referee comments, we found an error in the calculation of the location of the strongest updraft. We corrected all our calculations and revised Fig. 6b, 9b, 12b, 14b and Tab. 4 in the revised version of the manuscript. The changes are small and do not affect the main messages of the manuscript. Further, Figure 15 was changed using an other fit method.

Best wishes

Bärbel Vogel

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## Author Comment to Referee #1

**ACP Discussions doi: 10.5194/acp-2020-552-RC1, (Editor - Gabriele Stiller), ‘Strong variability of the Asian Tropopause Aerosol Layer (ATAL) in August 2016 at the Himalayan foothills’ by Sreeharsha Hanumanthu et al.**

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We thank Referee #1 for the positive review and for important further guidance on how to revise our manuscript. Our reply to the reviewer comments is listed in detail below. Questions and comments of the referee are shown in italics. Passages from the revised version of the manuscript are shown in blue.

*Based on COBALD measurements in North India in 2016 August, the variability of the ATAL features is analyzed, and the source regions is simulated with trajectory model - CLaMS. Some interesting results are derived, such as the strong variability of the ATALs altitude, vertical extend, and aerosol backscatter intensity. Some important transport pathways are identified for different ATAL intensity, such as continental convection and maritime typhoon. The phenomena with no ATAL detected is puzzling. Overall, this manuscript is well written and is recommended to be published in ACP.*

### Minor issues:

1. P2L12: 17km → 18km

done

2. P2L19-20: *The Asian summer monsoon circulation is affected first by the land-sea contrast, and second by the presence of the Tibetan Plateau. Therefore, this sentence should be modified.*

We agree and modified the sentence in the revises version of the manuscript as follows:

The dynamics and thermodynamics of the Asian monsoon are caused by the land–sea contrast and are influenced by the orography of the Himalayas and the adjacent mountain ranges (e.g. Turner and Annamalai, 2012, and references therein).

3. *P2L21: The monsoon anticyclone is linked to deep convection in summer over the Indian subcontinent AND OTHER ASIAN MONSOON REGIONS.*

Many thanks for this advice. We revised the sentence as follows in the revised version of the paper:

The Asian monsoon anticyclone is linked to deep convection in summer over south Asia and the associated diabatic heating (Hoskins and Rodwell, 1995; Randel and Park, 2006).

4. *P3L18-20: The Asian tropopause transition layer in summer is first investigated by Pan et al. (2014). Pan, L. L., L. C. Paulik, S. B. Honomichl, L. A. Munchak, J. Bian, H. B. Selkirk, and H. Vömel, 2014: Identification of the tropical tropopause transition layer using the ozone-water vapor relationship, J. Geophys. Res. Atmos., 119, doi:10.1002/2013JD020558.*

We added the following sentences regarding the Asian tropopause transition layer (ATTL) in the revised version of the paper to the introduction:

Pan et al. (2014) identified the tropical tropopause transition layer (TTL) using chemical tracer-tracer relationships in the tropics and over the Asian monsoon. Their comparison shows that the tracer-identified transition layer over the Asian monsoon is similar to the TTL, although the ATTL is located at higher potential temperature levels. However, during the monsoon season the vertical upward transport caused by radiative heating at the top the Asian monsoon anticyclone is faster than elsewhere in the tropics (Vogel et al., 2019).

5. *P4L1-2: How to show the size, radius or diameter?*

The reviewer raised a very relevant point here. In the revised version of the paper we give more details about size distribution of ATAL particles as follows:

Only few measurements of size distributions of particles in the ATAL are available, but measurements with an optical particle counter (OPC) (Deshler et al., 2003) from Hyderabad, India and measurements with a Printed Optical Particle Spectrometer (POPS) (Gao et al., 2016) from Kunming, China, (both in 2015) confirm the presence of the ATAL (Yu et al., 2017; Vernier et al., 2018). The OPC measurements indicate that the concentration of ATAL particles is highest close to the cold point tropopause and decline towards greater altitudes; concentrations (up to about 25 particles per  $\text{cm}^{-3}$ ) are dominated by particles in the  $r > 0.094 \mu\text{m}$  channel, whereas particle number concentrations for the  $r > 0.15 \mu\text{m}$  channel, and the  $r > 0.30 \mu\text{m}$  channel are lower by a factor of 30 and 300, respectively (Vernier et al., 2018, Fig. 9).

6. *P4L3-4: 0.6W/m<sup>2</sup> and 0.5K???*

Many thanks for this important advice. We revised the paragraph as follows in the revised version of the paper:

Based on CALIOP measurements, the summertime aerosol optical depth over Asia associated with the ATAL has increased from  $\approx 0.002$  to 0.006 between 1995 and 2013, resulting in a short-term regional forcing at the top of the atmosphere of  $-0.1 \text{ W/m}^2$  – compensating about one third of the comparable radiative forcing associated with the global increase in  $\text{CO}_2$  (Vernier et al., 2015). The regional radiative forcing caused by the ATAL, differs for clear and total sky conditions; total sky calculations show less shortwave radiative forcing over the monsoon region because of cloudiness (Vernier et al., 2015).

It is likely that, over Asia in the past  $\sim 20$  years, the altered radiative forcing has led to summertime reductions in surface temperature, although this effect is not quantified yet. However, the radiative forcing caused by the ATAL could be compared with the global aerosol forcing caused by moderate volcanic eruptions since 2000, which translates into a surface cooling of 0.05 to 0.12 K (Ridley et al., 2014).



7. *P4L7-10: In some ATAL studies, volcanic eruptions are removed, because volcanic signal is much stronger than ATAL, which will mask the effect of ATAL.*

Following the reviewer's advice we revised this paragraph in the revised version of the paper as follows:

The co-occurrence of the ATAL with volcanic eruptions in the region further enhances the radiative forcing by the ATAL; Fairlie et al. (2014, Fig. 8), based on monthly accumulations of CALIOP aerosol data between 14 and 40 km altitude, reported a top of the atmosphere radiative forcing locally over Asia and Europe during July 2011 in response to the Nabro volcanic eruption of about  $-0.8$  to  $-1.5$  W/m<sup>2</sup>. Therefore, periods of volcanic eruptions are removed in some ATAL studies; because the volcanic signal is much stronger than the ATAL signal itself, a volcanic signal will mask the ATAL (e.g. Thomason and Vernier, 2013; Vernier et al., 2015).

The impact of the ATAL is furthermore modulated by El Niño. During El Niño, the ATAL is thicker and broader over the Indian region, resulting in a reduction of the solar flux and a surface cooling of about 1 K over North India. An elevated ATAL over South Asia exacerbates the severity of Indian droughts (Fadnavis et al., 2019).

8. *P4L28-29: Nitrate aerosol is dominant in the ATAL is first suggested by Gu et al. (2016) by simulation. Gu, Y., H. Liao, and J. Bian, 2016: Summer-time nitrate aerosol in the upper troposphere and lower stratosphere over the Tibetan Plateau and the South Asian summer monsoon region, Atmos. Chem. Phys., 16, 6641-6663, doi:10.5194/acp-16-6641-2016.*

Many thanks for this advice. We revised the paragraph as follows in the revised version of the paper:

Only limited information on the chemical composition of the ATAL particles is available from measurements so far. From simulations, there is evidence that desert dust is lifted to UTLS altitudes and entrained into the ATAL (Fadnavis et al., 2013; Lau et al., 2018; Yuan et al., 2019). Further, simulations show elevated aerosol concentrations of sulphate, nitrate, ammonium, black carbon and organic carbon within the ATAL with contributions of nitrate dominating (Gu et al., 2016; Fairlie et al., 2020). Moreover,

Fairlie et al. (2020) found in model simulations for summer 2013 a different chemical compositions of the ATAL depending on the location within the Asian monsoon anticyclone with nitrate aerosol as the dominant component on the southern flank of the anticyclone.

9. *P6L30:  $CI > 7.0$ ,  $BSR_{940} > 2$  AND  $S_{ice} > 70\%$   $\rightarrow$  OR*

We have replaced the following text in the ACPD version:

‘We reject layers with  $CI > 7.0$ ,  $BSR_{940} \geq 2$  and  $S_{ice} > 70\%$  as cirrus clouds (Vernier et al., 2015; Li et al., 2018; Brunamonti et al., 2018)....’

in the revised version of the paper with:

We reject layers showing a  $CI > 7.0$ , and a  $BSR_{940} \geq 2$  as well as an enhanced ice saturation ( $S_{ice} > 70\%$ ) as cirrus clouds; clearly the CI criterion matters most for the detection of cirrus particles (Vernier et al., 2015; Li et al., 2018; Brunamonti et al., 2018).

10. *P6L30: Aerosol layers without cirrus can exist under the condition  $S_{ice} > 70\%$ .*

We agree, see text change above item 9.

11. *P6L32: Could you provide the vertical range of UTLS?*

We replaced ‘UTLS’ by the altitude range as follows:

Other sections of the profiles measured during the August soundings, which show substantially elevated values of  $BSR_{455}$  between  $\sim 140$  and  $70$  hPa ( $\approx 14$ – $18$  km), are classified as ATAL.

12. *P9L11-14: This ‘somewhat different picture’ can be explained by the results from Pan et al. (2014) as mentioned above. The distribution of lapse-rate minimum levels is compact in potential temperature scale but diffuse in the*

*altitude scale.*

In Pan et al. (2014), it is described that the distribution of the level of minimum stability (LMS) is more compact in potential temperature space and that the cold point tropopause (CPT) levels are more compact in the altitude space. This is consistent with the behaviour of temperature and potential temperature profiles in these two regions – LMS is the region of near constant potential temperature so the minimum gradient in this region can potentially vary over a large altitude range. The CPT has a very well defined altitude from the temperature profiles, but it is a region where potential temperature has a large gradient. However, in our work the distributions of ATAL with pressure and potential temperature as the vertical coordinate are discussed. It is difficult to directly link the distributions of the levels of CPT and LMS to the ATAL.

We revised this sentence as follows:

Using the backscatter measurements with potential temperature as the altitude scale, a somewhat different picture emerges than when using pressure as the altitude scale because the potential temperature has a large gradient in this altitude region.

13. *10L1: This result can also explained by the results on CPT from Pan et al. (2014).*

See above item 12.

14. *P18L1: The two branches are not easily found in the figure, could you show more clearly?*

We revised the description of the two branches to make the difference more clear in revised version of the manuscript as follows:

Focusing on the trajectories from the BL (Fig. 8; in the ACPD Version) two branches, a western and eastern branch, of trajectories are found depending on the altitude of the measurements (Fig. 7a; in the ACPD Version); trajectories from above 370 K are from eastern and trajectories from below 370 K

are from western longitudes. In the western branch of the trajectories the air masses are transported around the western mode of Asian monsoon anticyclone, while air masses from the eastern branch are coming from the Pacific ocean and are transported along the southern edge of the eastern mode of the anticyclone to the measurement location in Nainital.

15. *P27L1-6: Why post-monsoon cases are used as background signal for aerosols in the UTLS? Yes, there's no ATAL during winter. But, the general circulation is quite different for summer and winter, so obviously the source regions are different. Could you use cases with no ATAL during summer as background signal?*

We agree that the general circulation in winter is very different to summer conditions and therefore the source regions are different for air parcels in the UTLS over Asia during summer and winter. Therefore the enhancement in backscatter ratio ( $BSR_{455}$ ) in August compared to the November measurements is a good measure to infer the strength of the ATAL in summer in relation to conditions in winter, when more unpolluted marine air masses are transported to the UTLS over Asia. Therefore we use the November cases as background signal.

It would be also interesting to use 'no ATAL cases' during summer as background for further analysis however our study is focused on the COBALD measurements in Nainital 2016 which is a small statistical data base of 15 valid flights in August and two during November. In this data base, there are only two 'no ATAL cases' during summer both overlaid by a cirrus cloud. Therefore, we consider it premature using these two 'no ATAL cases' as the background signal.

16. *P27: For case 2, beside source regions, other parameters impacting ATAL such as temperature and  $S_{ice}$  should also be considered.*

We agree and added the following sentence:

However, we can not exclude that also aerosol removal processes depending on temperature and  $S_{ice}$  such as in-situ cirrus formation contribute to the

existence of the no ATAL measurement on 15 August 2016.

17. *P28L26: Some in situ measurements show that the aerosols concentration in the middle troposphere is very low, which should be considered in the argument.*

We agree that the aerosols concentration in the middle troposphere is very low in some in situ measurements which is also visible in the measurements in Nainital presented in this work (see Fig. 2 in the ACPD version). In the paper, we argue that the uplift (convection) of air masses from the lower troposphere could transport enhanced concentrations of aerosol particles to ATAL altitudes which is independent from the low aerosol concentration in the middle troposphere. This is indeed consistent with the OPC measurements presented by Vernier et al. (2018) that found much lower aerosol concentrations in the middle troposphere than at the ground or in the ATAL. We have now mentioned this point in the paper explicitly.

We have replaced the text in the ACPD version:

‘...however the uplift (convection) of air masses from the lower troposphere could transport enhanced concentrations of aerosol particles to ATAL altitudes (as proposed by Vernier et al. (2018)). Vernier et al. (2018) found in some balloon-borne measurements of the ATAL from Hyderabad, India, in summer 2015 that the presence of cirrus is associated ...’

in the revised version of the paper with:

...however the uplift (convection) of air masses from the lower troposphere could transport enhanced concentrations of aerosol particles from close to the ground to ATAL altitudes without much influence of aerosol in the middle troposphere (see Fig. 2 in the ACPD). This concept is consistent with the results of Vernier et al. (2018), who found in OPC balloon-borne measurements of the ATAL from Hyderabad, India, in summer 2015 that aerosol concentrations and in the ATAL are of the same magnitude, while much lower aerosol concentrations prevail in the middle troposphere. In their balloon measurements Vernier et al. (2018) further found that the presence of cirrus is associated ...

18. *P31L24-26: Factors impacting the variability of the ATAL include not only source regions, but also other parameters related the formation and growth of aerosol, the latter should also be considered.*

We agree and added the reviewer's argument to the revised version of the manuscript as follows:

In this work, we show that there is a variety of factors impacting the variability of the ATAL: continental convection, tropical cyclones (maritime convection), dynamics of the anticyclone and stratospheric intrusions. All these factors contribute to the observed day-to-day variability of the ATAL found over Nainital in August 2016. The ATAL is also impacted by other processes related to the formation and growth of aerosol as well as by aerosol removal processes such as in-situ cirrus formation.

and add the following text to the second paragraph of the conclusions:

... However, we have only one no ATAL measurement in August 2016 (partly influenced by cirrus), therefore we can not exclude that also aerosol removal processes such as in-situ cirrus formation contribute to the existence of no ATAL measurements. More balloon-borne measurements would be required to validate this hypothesis.

19. *P32L1-4: Possibly, satellite data for aerosol can be used.*

Many thanks for the reviewer's suggestion, however the strong day-to-day variability of the altitude, the vertical extent and the backscatter intensity of the ATAL at UTLS altitudes is not visible in the climatological mean values of the ATAL derived by satellite observations. Therefore we are not sure if satellite data could really help to prove our hypothesis that the main driver for 'no ATAL cases' are the dynamics of the Asian monsoon anticyclone. In any event, an in-depth study of the averaging procedure necessary to derive ATAL information from satellite observations would be required for such an analysis. This is more than what we can achieve in this paper. Furthermore, a larger data base of high-resolution measurements (e.g. from balloons or high-altitude aircraft) would also be very helpful to get a deeper understanding of 'no ATAL cases' during Asian summer monsoon.

20. *P32L20-22: I think its too early to talk about the regulations, because we still dont know whether the ATAL existence is good or bad to human beings.*

We agree and discuss this issue more controversially in the revised version of the paper by adding the following sentence:

On the other hand, the ATAL impacts the radiative balance of the Earth's atmosphere and could have potential positive side effects in terms of reducing surface temperatures (Fadnavis et al., 2019), therefore more future research about the ATAL and its impacts is required.

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## Author Comment to Referee #2

**ACP Discussions doi: 10.5194/acp-2020-552-RC1, (Editor - Gabriele Stiller), ‘Strong variability of the Asian Tropopause Aerosol Layer (ATAL) in August 2016 at the Himalayan foothills’ by Sreeharsha Hanumanthu et al.**

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We thank Referee #2 for important further guidance on how to revise our manuscript. Our reply to the reviewer comments is listed in detail below. Questions and comments of the referee are shown in italics. Passages from the revised version of the manuscript are shown in blue.

*Understanding the nature, origin and impacts of the Asian Tropopause Aerosol Layer has been a research focus for nearly a decade. Recent airborne campaigns conducted in Asia during the Summer Monsoons have provided a wealth of information about the ATAL that are rapidly advancing our understanding of this phenomenon. As a part of the StratoClim field experiment that took place in Nepal and India in 2017, this study present results from the balloon flights conducted from Nainital in August 2017 compared with those obtained in November 2016. The balloon flights reveal an imporant day-to-day variability of the Scattering Ratio (SR) taken as a relative measure of aerosol loadings. In order to understand the causes of those fluctuations, the authors run the CLAMS trajectory model to distinguish the origin of air masses in the Boundary Layer, the free troposphere and the lower stratosphere from different part of Asia. They concluded that large SR values within the ATAL tend to be associated with air masses from the Tibetan plateau, Himalayan foothills and lands while oceanic origin tend to result in a depletion of UTLS aerosols.*

*Despite some grammatical mistakes and relatively lengthy manuscript, which could be, shorten and better summarize, this is an interesting study, which merits its publication in ACP. However, I suggest significant revisions to make this possible. Because deep convection is a fundamental transport pathways for air mass to move from the Boundary Layer into the Upper Troposphere and Lower Stratosphere during the Monsoon, the coarse resolution of the meteorological field used to run CLAMS likely result in misrepresentation of the vertical transport pathways*

*especially after a few days when the likelihood of encountering deep convection is very high. The manuscript lacks a deeper analysis of the role of convective storms that influence the vertical transport of air masses and those measurements. Other studies have used Cloud Top Temperature as a proxy for deep convection to find out the location where air masses are influenced by deep convection and I believe that this study would need to adopt a similar approach to be more convincing. Below are additional technical comments of this paper that the authors may want to consider.*

The reviewer's major comment regarding the representation of convection within CLaMS back-trajectory calculations driven by ERA-Interim reanalysis is discussed in detail under item # 5. We agree that the manuscript is somewhat long therefore we followed the reviewer's advice and removed Fig. 14b, c as well as Fig. 15b (ACPD version) and the corresponding text in Sect. 4.4. Further, the manuscript was carefully proof-read regarding to grammatical mistakes by several of the authors. To avoid any misunderstanding we would like to point out that the paper contains only balloon-borne measurements from India (Nainital) in 2016. Balloon-borne measurements were also performed in Nepal in summer 2017 in the frame of the StratoClim project, however no COBALD measurements are available for 2017.

## **Minor issues:**

1. *Title. Im not sure the title translate very well the topics of this paper. Moreover, the term strong variability is confusing if not related to time information (in this case day-to-day or intraseasonal variability).*

We agree and changed the title from

‘Strong variability of the Asian Tropopause Aerosol Layer (ATAL) in August 2016 at the Himalayan foothills’ to

[Strong day-to-day variability of the Asian Tropopause Aerosol Layer \(ATAL\) in August 2016 at the Himalayan foothills](#)

2. *P1/L3. I believe that ‘inside’ is not required. Its understood from the previous part of the sentence.*

We prefer to keep 'inside' to avoid any misunderstanding.

3. *P1/L7. 'COBALD' does not need to be repeated here. It could be replaced by 'compared to those obtained...'*

We prefer to keep 'COBALD' to have a clear message.

4. *P1/L12-L13. Not 'composition' but scattering ratio.*

Thanks for the comment. We revised the sentence

'We identify the transport pathways of air parcels contributing to the ATAL over Nainital in August 2016, as well as the source regions of the air masses contributing to the composition of the ATAL.'

in the revised version of the paper as follows:

We identify the transport pathways as well as the source regions of air parcels contributing to the ATAL over Nainital in August 2016.

5. *P1/L18-21. This is related to the major comment I have on this study. How realistic is the vertical transport pathway described here relative to direct injection by deep convection ?*

Within the Nainital measurements in summer 2016 the ATAL is located from 360 K up to 420 K. The top of the convective outflow level is around 360 K. Deep convection events are most likely underestimated in ERA-Interim. The new high-resolution ECMWFs next-generation reanalysis ERA5 (Hoffmann et al., 2019; Hersbach et al., 2020) has much higher spatial and temporal resolution than ERA-Interim. A recent comparison between CLaMS trajectories driven by ERA-Interim and by ERA5 investigated the impact of tropical cyclones on ozone and water vapour measured during the SWOP balloon-campaign in 2009 and 2015 in Kunming (China) within the Asian monsoon anticyclone (Li et al., 2020). Different vertical transport via deep convection is found depending on the employed reanalysis data

(ERA-Interim, ERA5) and vertical velocities (diabatic, kinematic). Both the kinematic and the diabatic trajectory calculations using ERA5 data show faster and stronger vertical transport than ERA-Interim primarily due to ERA5's better spatial and temporal resolution, likely resolving more convective events. Although the details of the vertical transport are different in all the cases studied by Li et al. (2020), the convective upward transport by tropical cyclones is found in ERA-Interim (diabatic) as well as in ERA5 (diabatic and kinematic). The location of the convective updraft is compared with brightness temperature from IR channel of FY-2D satellite and with cloud top temperature from FY-2G satellite showing that ERA-Interim (diabatic) as well as ERA5 (diabatic, kinematic) trajectories have the convective upward transport in the region of the tropical cyclone, whereby ERA5 (diabatic) fits best with the center of the cyclone. Further, the transport pathway above the convective outflow level driven by both the anticyclonic flow and diabatic heating in the region of the Asian monsoon anticyclone ('upward spiralling range') is found in both reanalysis data (ERA-Interim, ERA5). Li et al. (2020) show that low ozone and low water vapour mixing ratios near the tropopause measured in August 2009 and 2015 in Kunming are the result of the interplay between the uplift of dry ozone-poor maritime air within tropical cyclones and the transport in the UTLS driven by the Asian monsoon anticyclone.

We are aware that using ERA5 for CLaMS trajectory calculations for the ATAL measurements in Nainital 2016 would yield deeper insights into the impact of deep convection to ATAL, however a study using ERA5 goes beyond the scope of the study presented here. Further studies are prerequisite to validate convection and diabatic heating rates in ERA5. In this study, we highlight the day-to-day variability of ATAL during August 2016 and its relation to continental and maritime convection. The use of high-resolution meteorological data and the use of satellite measurements (e.g. Cloud Top Temperature) would be necessary to identify the detailed location of single convection events. However, the results by Li et al. (2020) encourage us that the representation of convection in ERA-Interim is adequate for the study presented here.

6. *P2/L5. Chinese emissions have decreased drastically over the past 2 decades so Sulfur emission in Asia are overall on the decreasing side so this sentence needs to be modified.*

We agree with the reviewer's comment and revised the sentence

‘Because of the strong growth of Asian economies, increasing anthropogenic emissions in the future are expected to enhance the thickness and intensity of the ATAL, thereby also enhancing the global stratospheric aerosol loading, which likely impacts surface climate.’

in the revised version of the paper as follows:

On the one hand increasing anthropogenic emissions in the future are expected due to the strong growth of Asian economies, on the other hand implementation of new emission control measures (in particular in China) have reduced substantially the anthropogenic emissions of some pollutants contributing to the ATAL. It needs to be monitored in the future, whether the thickness and intensity of the ATAL will further increase, which likely impacts surface climate.

Further, Chinese emissions are discussed in the Introduction of the ACPD version as well (P3, L25-27).

7. P2/L9- L11. *I would argue that Chinese balloon-borne measurements and ground-based lidar suggested the presence of aerosol layers over the Tibetan Plateau earlier than satellite observations but the extension of the ATAL was indeed discovered through global satellite observations. Refs.:*

Kim, Y.-S., T. Shibata, Y. Iwasaka, G. Shi, X. Zhou, K. Tamura, and T. Ohashi, 2003: *Enhancements of aerosols near the cold tropopause in summer over Tibetan Plateau: Lidar and balloonborne measurements in 1999 at Lhasa, Tibet, China. Lidar Remote Sensing for Industry and Environment Monitoring III*, U. N. Singh, T. Itabe, and Z. Liu, Eds., *Society of Photo-Optical Instrumentation Engineers (SPIE Proceedings, Vol. 4893)*, 496503, <https://doi.org/10.1117/12.466090>.

Tobo, Y., Y. Iwasaka, G.-Y. Shi, Y.-S. Kim, T. Ohashi, K. Tamura, and D. Zhang, 2007: *Balloon-borne observations of high aerosol concentrations near the summertime tropopause over the Tibetan Plateau. Atmos. Res.*, 84, 233241

We thank the reviewer for these references; we were indeed not aware of these measurements. As suggested the measurements in August 1999 are now mentioned in the paper, when the ATAL is introduced. We added the following text:

Although the large horizontal extent of the ATAL was first seen in the CALIOP measurements (Vernier et al., 2011, 2015), first observations of enhanced number concentrations of sub-micron aerosol particles between 130–70 hPa in the Asian summer monsoon were made during a balloon ascent from Lhasa (29.7°N, 91.1°E) already in August 1999 (Kim et al., 2003; Tobo et al., 2007).

And we are now also referring to the papers in question in the discussion of measurements of size distributions of particles in the ATAL in the introduction further below:

Only few measurements of size distributions of particles in the ATAL are available. Early size-resolved measurements using a balloon-borne optical particle counter (OPC) in August 1999 showed high number concentrations (0.7–0.8 particles  $\text{cm}^{-3}$ ) of aerosol particles with radii of 0.15–0.6  $\mu\text{m}$  between about 130–70 hPa in the Asian summer monsoon (Kim et al., 2003; Tobo et al., 2007).

8. *P2/L17. This sentence needs to be more accurately stated. The paper did not suggest the presence of the ATAL in the 90s but the presence of ammonium nitrate and since we do not know the overall contribution of AN within the ATAL, It's hard to formalize a general statement such as the one here. I suggest being more accurate.*

The paper by Höpfner et al. (2019) contains the following statement: “The spatially resolved AN observations with the CRISTA satellite reveal that enhanced concentrations of AN (0.05–0.3  $\mu\text{g m}^{-3}$ ) are located only within the AMA (Fig. 1). These observations between 8 and 16 August 1997 indicate that an ATAL was present in the Asian monsoon UT in summer 1997, years earlier than hitherto thought”.

However, we agree that a more careful wording is appropriate here; we have changed the part of the sentence starting with “although” by:

However, Höpfner et al. (2019) reported that as early as 1997, during the Asian monsoon period, enhanced concentrations of solid ammonium nitrate



particles were present throughout the Asian monsoon anticyclone.

9. *P2/L26-27. I would suggest targeting the citations that are most appropriate for this statement.*

Many thanks for this comment. We agree that a plenty of citations are included for this statement. However, in these citations different measurements and chemical species are used. Therefore, we think it is useful to cite all of them here.

10. *P3/L12. I would suggest being quantitative in this sentence. What are the contributions from India and China?*

We agree and added the percentages from India and China to the revised version of the paper as follows:

Source apportionment of the model simulations by Fairlie et al. (2020) indicated the dominance of the contribution of regional anthropogenic emissions from China and the Indian subcontinent (both  $\sim 30\%$  attributable to regional  $\text{SO}_2$  sources) to aerosol concentrations in the ATAL in August 2013.

In addition, the Chinese  $\text{SO}_2$  emissions (Zheng et al., 2018) are also discussed in the Introduction of the ACPD version (P3, L25-27).

11. *P3/L21. Could you explain why the results seem to be consistent with Brunamonti et al., 2018 ?*

Brunamonti et al. (2018) denote the altitude range between maximum convective outflow and the cold point tropopause as the Asian tropopause transition layer (ATTTL). Above, the ATTTL, based on  $\text{H}_2\text{O}$  measurements and trajectory calculations, Brunamonti et al. (2018) found a layer of a confined air in the lower stratosphere.

Vogel et al. (2019) denote the region where air masses are uplifted by diabatic heating across the (lapse rate) tropopause from about 360 K up to 460 K as the ‘upward spiralling range’. The higher the air masses are above the thermal tropopause, the larger the contribution of air masses is from

outside the Asian monsoon anticyclone from the stratospheric background coming into the upward spiralling flow.

Both concepts explain the confinement of air in the Asian monsoon anticyclone directly above the cold point tropopause. The higher the air masses are above the thermal tropopause, the weaker is the confinement that can be explained by the larger contribution of air masses from the stratospheric background. This yields a vertical profile of ATAL as shown in Fig. 1 (right; in the ACPD Version of the paper). In Fig. 1 (right), the potential temperature is considered as the vertical coordinate, a top of the ATAL is not clearly defined. Only a slow decrease of averaged  $BSR_{455}$  with altitude is observed. This observation is consistent with a decreasing confinement of the air mass within the Asian monsoon anticyclone with increasing potential temperature (Brunamonti et al., 2018; Vogel et al., 2019).

12. *P4/L4. Is there a reference for those estimates?*

Many thanks for this important advice. We revised the paragraph as follows in the revised version of the paper:

Based on CALIOP measurements, the summertime aerosol optical depth over Asia associated with the ATAL has increased from  $\approx 0.002$  to 0.006 between 1995 and 2013, resulting in a short-term regional forcing at the top of the atmosphere of  $-0.1 \text{ W/m}^2$  – compensating about one third of the comparable radiative forcing associated with the global increase in  $\text{CO}_2$  (Vernier et al., 2015). The regional radiative forcing caused by the ATAL, differs for clear and total sky conditions; total sky calculations show less shortwave radiative forcing over the monsoon region because of cloudiness (Vernier et al., 2015).

It is likely that, over Asia in the past  $\sim 20$  years, the altered radiative forcing has led to summertime reductions in surface temperature, although this effect is not quantified yet. However, the radiative forcing caused by the ATAL could be compared with the global aerosol forcing caused by moderate volcanic eruptions since 2000, which translates into a surface cooling of 0.05 to 0.12 K (Ridley et al., 2014).

13. *Overall, the introduction could be improved by organizing the different*

*paragraph with titles.*

Caused by the comments of Reviewer #1 and #2 we carefully revised the introducing in several places. Therefore, the introduction in the revised version is somewhat longer than the ACPD version in contrast to the Reviewer's advice to shorten the manuscript. We grouped related topics in different paragraphs without using subtitles.

14. P5/L9. *A reference to Pandit et al., 2015 could be added here . Ref: Pandit, A. K., Gadhavi, H. S., Venkat Ratnam, M., Raghunath, K., Rao, S. V. B., and Jayaraman, A.: Long-term trend analysis and climatology of tropical cirrus clouds using 16 years of lidar data set over Southern India, Atmos. Chem. Phys., 15, 13833-13848, <https://doi.org/10.5194/acp-15-13833-2015>, 2015.*

We agree and have added Pandit et al. (2015) at P5/L9 (ACPD Version).

15. P6/L4. *A calibration adjustment is needed to fit the COBALD raw signal to the molecular scattering. A few lines describing a little better the procedure could be added here.*

Many thanks for this comment. We have replaced the following text in the ACPD version

‘The ATAL analysis is mainly based on the COBALD 455 nm measurements, in particular on the backscatter ratio (BSR), which is defined as the ratio of the COBALD raw signal (from particulates and air molecules) over the pure molecular scattering (derived from the ambient molecular number density, using the temperature and pressure measured by RS41).’

in the revised version of the paper with:

The ATAL analysis is mainly based on the COBALD 455 nm measurements. The COBALD data are expressed as backscatter ratio (BSR), i.e., the ratio of the total-to-molecular backscatter coefficient. This is calculated by dividing the total measured signal by its molecular contribution, which is computed from the atmospheric extinction according to Bucholtz (1995), and using air density derived from the RS41 temperature and pressure measurements.

16. *P6L16. If Im not mistaken  $IST = UTC + 5h30$ ...(not 6h).*

We agree that  $IST = UTC + 5h30$ . We corrected that in the revised version of the manuscript as follows:

All balloon soundings with a COBALD instrument were launched at night-time between 23:00 Indian Standard Time (IST) (corresponding to Coordinated Universal Time (UTC) +5:30 h) and next day 03:00 IST.

and also corrected the figure caption of Fig. 3 (ACPD Version):

Location of the Asian monsoon anticyclone measured by the geopotential height at 110 hPa for 6 August, 15 August and 18 August 2016 at 18:00 UTC (corresponding to 23:30 local time (IST) in Nainital).

17. *P6.L30. You probably mean to say ‘we identify ice clouds with...’*

We have replaced the following text in the ACPD version including also the comments of Reviewer #1:

‘We reject layers with  $CI > 7.0$ ,  $BSR_{940} \geq 2$  and  $S_{ice} > 70\%$  as cirrus clouds (Vernier et al., 2015; Li et al., 2018; Brunamonti et al., 2018)....’

in the revised version of the paper with:

We define clear-sky (i.e. cloud-free) conditions; only layers are used in the ATAL analysis where the criteria  $CI < 7$ ,  $BSR_{940} < 2$  and  $S_{ice} < 70\%$  are simultaneously fulfilled. All other measurements are rejected from the ATAL analysis. The 70% threshold is on purpose low enough, that it ensures no cirrus cloud measurements are misinterpreted as aerosol measurements (Vernier et al., 2015; Li et al., 2018; Brunamonti et al., 2018).

18. *P9L14. Aerosol scavenging also depends on aerosol size and composition, which affect their ability to uptake water.*

We have replaced the following text in the ACPD version:

‘This steep gradient represents the top of the convective outflow region (e.g., Gettelman and de Forster, 2002), i.e. below this level more frequent deep convection scavenges the aerosol.’

in the revised version of the paper with:

This steep gradient represents the top of the convective outflow region (e.g., Gettelman and de Forster, 2002), i.e. below this level aerosols are more frequently scavenged and removed by precipitation.

19. *P13L19. How trustable are trajectories run beyond a week ?*

We added the following paragraph to Sect. 2.4 (Trajectory calculations):

In general, trajectory calculations have limitations due to trajectory dispersion depending on the trajectory length. However, the frequently employed trajectory length to study transport processes in the Asian monsoon region is ranging from a couple of weeks to a few months depending on the transport times from Earth's surface to atmospheric altitudes (e.g. Chen et al., 2012; Bergman et al., 2013; Vogel et al., 2014; Garny and Randel, 2016; Müller et al., 2016; Li et al., 2017, 2018). The CLaMS backward calculations to analyse the balloon measurements in Nainital show that the shortest transport times from Earth's boundary layer to ATAL altitudes ( $\sim 360\text{--}420\text{ K}$ , see Tab. 1; ACPD Version) is about 10-15 days (see Sect. 4; ACPD Version). Only a few trajectories are shorter. However, most of the trajectories show longer transport times than two weeks to reach ATAL altitudes (up to  $\sim 420\text{ K}$  or  $\sim 75\text{ hPa}$ ). The vertical upward transport of air parcels above the maximum level of convective outflow ( $\sim 360\text{ K}$ ) is determined by diabatic heating rates which are up to  $\sim 1\text{--}1.5\text{ K}$  per day over the region of the Asian monsoon anticyclone during summer based on ERA-Interim (Vogel et al., 2019). Convection occurs on short time scales of hours to a few days, whereby the transport from  $360\text{ K}$  up to  $420\text{ K}$  needs 40 to 60 days using ERA-Interim. Therefore we performed 40-day backward trajectory calculations here, as well as trajectories with a length of 60 and 80 days to test the sensitivity on the trajectory length.

20. *P14. Table 2 needs to be better explained. What's the definition of the variable in the table? Residence time in a given layer relative to the sum?*

We added in the revised version of the paper the following lines to the caption of Fig. 2:

In the second and third row, the selection criteria for BL, LT, UT, and LS are listed. Trajectories are considered ending in the BL, when they are located for the first time below about 2–3 km (i.e., hybrid coordinate  $\zeta \leq 120$  K). The location of this point is referred to as ‘end point’ of the trajectory in the model boundary layer. For the remaining trajectories ending at atmospheric altitudes ( $\zeta > 120$  K), a potential temperature criterion ( $\Theta$ ) is employed to discriminate between origins in LT, UT and LS.

The parameters within Tab. 2 are also explained within Sect. 4 (P13 L29 – P15 L3; ACPD Version).

21. *P30. Figure 15. Why do you choose to take the mean value? I would suggest to plot the same with the value corresponding to the altitude where the model was initialized.*

It seems that there is a misunderstanding. In Fig. 15 (ACPD version) the mean value of the backscatter intensity ( $\overline{\text{BSR}}_{455}$ ) between bottom and top of ATAL is shown calculated for each balloon sounding using the binned data (see Tab. 1; ACPD version) as explained in Sect. 3.1. Cirrus clouds between top and bottom are excluded in the calculation of  $\overline{\text{BSR}}_{455}$ . Therefore,  $\overline{\text{BSR}}_{455}$  is the backscatter intensity averaged over the ATAL altitude range listed in Tab. 1 for each flight.

We added the following sentence to the figure caption of Fig. 15 (ACPD version) to the revised version of the paper

$\overline{\text{BSR}}_{455}$  is the backscatter intensity averaged over the ATAL altitude range listed in Tab. 1 for each flight (details see Sect. 3.1; ACPD version).

22. *Figure 15. What are the correlation coefficient values? Im not sure if you can draw much conclusions from this plot apart overall tendency.*

Many thanks for this comment. We added a new table (Tab. 1 within this authors’ reply; Tab. 5 in the revised version) showing the correlation coefficients for each region and added the following paragraph to the revised version of the manuscript:

Further, the Pearson correlation coefficients between the different regions (Tibet, Foothills, Land and Ocean) and the backscatter intensity averaged

over the ATAL altitude range ( $(\overline{\text{BSR}}_{455}-1) \times 100$ ) for the location of the end points of the trajectories and for the location of the strongest updraft along the trajectories is calculated (see Tab. 1; in this author comment). Low positive correlations (i.e. values from 0.3 to 0.5) are found for the location of the end points of Tibet and Foothills as well as for the location of the strongest updraft of Tibet.

Moreover, the Pearson correlation coefficients are calculated excluding also the flights with a relatively low absolute number of trajectories (11, 21, 23 and 26 August) and the no ATAL case on 15 August. The no ATAL case is excluded because the low backscatter intensity is not completely explained through the backward trajectory analysis presented here. Moderate positive correlations (i.e. values from 0.5 to 0.7) are found for Tibet, Foothills and Land with the largest correlation coefficient of 0.7 for the location of endpoints in Tibet. Low negative correlations (i.e. values from  $-0.3$  to  $-0.5$ ) are found for contributions from the Ocean. Overall, our findings show that the stronger the backscatter intensity averaged over the ATAL altitude range, the higher the continental contributions from Tibet, Foothills and Land. The weaker the ATAL is during August 2016, the higher are the maritime contributions.

23. *P31/L17. This phrase needs to be nuanced. While it is true that the signal-to-noise ratio from the CALIPSO space-borne lidar does not allow studying day-to-day variability of the ATAL, observations from SAGE II/SAGEIII can be potentially used for that.*

Following the reviewer’s advice we revised the sentence

‘This variability is not visible in the climatological mean values of the ATAL derived by satellite observations.’

in the revised version as follows:

In contrast to high-resolution in situ measurements, space-borne observations often do not allow studying the day-to-day variability of ATAL profiles therefore mean values of ATAL profiles were frequently used in previous studies based on satellite observations. (e.g. Vernier et al., 2011, 2015).

24. *P31/L24. I don’t think the impact of convection, which is not well represented in ERA-Interim, has been fully explored and thus must bias most of*

Pearson Correlation Coefficients				
location of	Tibet	Foothills	Land	Ocean
end points	0.33	0.35	0.14	-0.19
strongest updraft	0.41	0.09	-0.00	-0.19
end points*	0.70	0.63	0.56	-0.27
strongest updraft*	0.69	0.52	0.58	-0.32

Table 1: Pearson correlation coefficients between the different regions (Tibet, Foothills, Land and Ocean) and the backscatter intensity averaged over the ATAL altitude range  $((\overline{\text{BSR}}_{455}-1) \times 100)$  for the location of the end points of the trajectories (see Fig. 15; revised version) and for the location of the strongest updraft. For each region the Pearson correlation coefficient was calculated using all measurements except the one on 12 August, when the UTLS is filled by a 5 km thick cirrus cloud. Further, the Pearson correlation coefficients are also calculated excluding in addition low statistics flights (11, 21, 23 and 26 August) where the trajectory number is lower than 50% of the maximum number of trajectories (#704) calculated on 18 August 2016 (see Fig. 14a; revised version) as well as the no ATAL case on 15 August (marked by \*).

*the trajectory results presented in this paper.*

Thank you for this comment. We added in the revised version of the paper, the following sentence to the conclusions

Clearly the details of the vertical transport will differ, when a higher resolution, more recent reanalysis data set would be used, however the general transport patterns will likely remain unaltered. For example, the convective upward transport by tropical cyclones is found consistently for ERA-Interim and ERA5 in diabatic calculations (Li et al., 2020).

and the following statement to Sect. 2.4 (Trajectory calculations):

In ERA-Interim changes are implemented to improve deep and mid-level convection compared to previous reanalysis data (Dee et al., 2011). However, small-scale rapid uplift in convective cores is not included. Therefore convection over Asia is most likely underestimated in ERA-Interim. Nevertheless, upward transport in larger convective systems such as tropical cyclones is well represented in CLaMS trajectory calculations driven by



ERA-Interim as shown in Li et al. (2017, 2020) by comparison with brightness and cloud top temperature derived from satellite observations.

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# Strong day-to-day variability of the Asian Tropopause Aerosol Layer (ATAL) in August 2016 at the Himalayan foothills

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## Abstract.

The South Asian summer monsoon is associated with a large-scale anticyclonic circulation in the Upper Troposphere and Lower Stratosphere (UTLS), which confines the air mass inside. During boreal summer, the confinement of this air mass leads to an accumulation of aerosol between about 13 km and 18 km (360 K and 440 K potential temperature), this accumulation of aerosol constitutes the Asian Tropopause Aerosol Layer (ATAL). We present balloon-borne aerosol backscatter measurements of the ATAL performed by the Compact Optical Backscatter Aerosol Detector (COBALD) instrument in Nainital in Northern India in August 2016, and compare these with COBALD measurements in the post-monsoon time in November 2016. The measurements demonstrate a strong variability of the ATAL's altitude, vertical extent, aerosol backscatter intensity and cirrus cloud occurrence frequency. Such a variability cannot be deduced from climatological means of the ATAL as they are derived from satellite measurements. To explain this observed variability we performed a Lagrangian back-trajectory analysis using the Chemical Lagrangian Model of the Stratosphere (CLaMS). We identify the transport pathways ~~of air parcels contributing to the ATAL over Nainital in August 2016,~~ as well as the source regions of ~~the air masses~~ air parcels contributing to the ~~composition of the ATAL.~~ ATAL over Nainital in August 2016. Our analysis reveals a variety of factors contributing to the observed day-to-day variability of the ATAL: continental convection, tropical cyclones (maritime convection), dynamics of the anticyclone and stratospheric intrusions. Thus, the air in the ATAL is a mixture of air masses coming from different atmospheric ~~height~~ altitude layers. In addition, contributions from the model boundary layer originate in different geographic source regions. The location

of strongest updraft along the backward trajectories reveal a cluster of strong upward transport at the southern edge of the Himalayan foothills. From the top of the convective outflow level (about 13 km; 360 K) the air parcels ascend slowly to ATAL altitudes within a large-scale upward spiral driven by the diabatic heating in the anticyclonic flow of the South Asian summer monsoon at UTLS altitudes. Cases with a strong ATAL typically show boundary layer contributions from the Tibetan Plateau, the foothills of the Himalayas and other continental regions below the Asian monsoon. Weaker ATAL cases show higher contributions from the maritime boundary layer, often related to tropical cyclones, indicating a mixing of ~~unpolluted and polluted air masses. Because of clean maritime and polluted continental air. On the one hand increasing anthropogenic emissions in the future are expected due to~~ the strong growth of Asian economies, ~~increasing anthropogenic emissions on the other hand implementation of new emission control measures (in particular in China) have reduced substantially the anthropogenic emissions of some pollutants contributing to the ATAL. It needs to be monitored~~ in the future ~~are expected to enhance~~, whether the thickness and intensity of the ATAL ~~, thereby also enhancing the global stratospheric aerosol loading will further increase~~, which likely impacts surface climate.

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## 1 Introduction

The existence of an Asian Tropopause Aerosol Layer (ATAL) in Northern hemisphere summer (June, July and August) was first discovered in lidar measurements of aerosol particles in the Upper Troposphere and Lower Stratosphere (UTLS) ~~performed by CALIPSO, by~~ the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation ~~(Vernier et al., 2011, 2015).~~ ~~(CALIPSO) mission (Vernier et al., 2011, 2015). CALIPSO is measuring the total attenuated backscatter through the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP).~~ The ATAL occurs between about 360 K and 440 K potential temperature (between about 13 and ~~17~~18 km); based on CALIOP measurements, the thickness of the ATAL is estimated to be 3–4 km between 30°N and 40°N and is thinner near the equator (Vernier et al., 2011, 2015, 2018; Bian et al., 2020). ~~Although the large horizontal extent of the ATAL was first seen in the CALIOP measurements (Vernier et al., 2011, 2015), first observations of enhanced number concentrations of sub-micron aerosol particles between 130-70 hPa in the Asian summer monsoon were made during a balloon ascent from Lhasa (29.7°N, 91.1°E) already in August 1999 (Kim et al., 2003; Tobo et al., 2007).~~ The existence of the ATAL is ~~also further~~ confirmed by in situ balloon-borne backscatter measurements (Vernier et al., 2015, 2018; Brunamonti et al., 2018) ~~in Lhasa (August 2013), Saudi Arabia (August 2015) and India (August 2016, 2017).~~ Solar occultation observations by the Stratospheric Aerosol and Gas Experiment (SAGE) II ~~(Thomason and Vernier, 2013)~~ ~~also confirmed the existence of the ATAL, but~~ indicated the absence of the ATAL prior to 1999 ~~, although a recent study suggested that an ATAL already existed in (Thomason and Vernier, 2013). However, Höpfner et al. (2019) reported that as early as 1997 (Höpfner et al., 2019), during the Asian monsoon period, enhanced concentrations of solid ammonium nitrate particles were present throughout the Asian monsoon anticyclone.~~

The existence of the ATAL is related to the presence of the Asian summer monsoon circulation and, in particular, to the Asian monsoon anticyclonic circulation at UTLS altitudes from about June to September. The ~~Asian summer monsoon circulation is affected by the presence of~~ dynamics and thermodynamics of the Asian monsoon are caused by the land-sea contrast and are influenced by the orography of the Himalayas and the adjacent mountain ranges (Boos and Kuang, 2010).  
 5 ~~The~~ (e.g. Turner and Annamalai, 2012, and references therein). The Asian monsoon anticyclone is linked to deep convection in summer over ~~the Indian subcontinent south~~ Asia and the associated diabatic heating (Hoskins and Rodwell, 1995; Randel and Park, 2006). Deep convection occurs primarily just south of and over the lower elevations of the Himalayan barrier; isolated deep convective events occur over the Tibetan Plateau (Houze Jr. et al., 2007). This deep convection in the region of the Asian monsoon anticyclone transports pollutants (e.g., CO, HCN, C<sub>2</sub>H<sub>6</sub>, SO<sub>2</sub>, NH<sub>3</sub>, HCFC-22) emitted at the surface in south  
 10 Asia to UTLS altitudes, where they are confined in the anticyclonic circulation in summer to a certain extent (e.g., Park et al., 2008; Randel et al., 2010; Chen et al., 2012; Ploeger et al., 2015; Chirkov et al., 2016; Garny and Randel, 2016; Santee et al., 2017; Höpfner et al., 2019; Bian et al., 2020).

The source regions of ATAL aerosols and their chemical precursors on the Earth's surface (origin) as well as the transport pathways from the surface to ATAL altitudes are poorly understood. The contributions of different boundary source regions  
 15 in Asia to the chemical composition of the Asian monsoon anticyclone are discussed in a variety of studies (e.g. Li et al., 2005; Park et al., 2009; Chen et al., 2012; Bergman et al., 2013; Fadnavis et al., 2014; Vogel et al., 2015; Pan et al., 2016; Li et al., 2017; Fairlie et al., 2020), identifying sources in India, the Tibetan Plateau, southwest China, southeast Asia, as well as from the Western Pacific. In addition, there is an intraseasonal change of boundary layer source regions contributing to the composition of the anticyclone that depends on the variability of convection and the spatial coverage of the anticyclone  
 20 itself (Vogel et al., 2015). Further in model simulations, it was found that during the monsoon peak season large quantities of dust are transported from the Middle East deserts to the upper troposphere and accumulate over the southern and eastern foothills of the Tibetan Plateau (Fadnavis et al., 2013; Lau et al., 2018). Lau et al. (2018) highlighted two key pathways lofting carbonaceous aerosols and dust by orographically forced deep convection, one over the Himalayas-Gangetic Plain (India) and another one over the Sichuan Basin (China). ~~Based~~ Further, based on aircraft measurements in summer 2017, Höpfner et al.  
 25 (2019) concluded that enhanced NH<sub>3</sub> concentrations (precursor of solid ammonium nitrate particles) over India were injected by strong convection into the upper troposphere over northwest India and northeast Pakistan. Source apportionment of the model simulations by Fairlie et al. (2020) indicated the dominance of the contribution of regional anthropogenic emissions from China and the Indian subcontinent (both ~ 30% attributable to regional SO<sub>2</sub> sources) to aerosol concentrations in the ATAL in August 2013.

Only limited information on the chemical composition of the ATAL particles is available from measurements so far. From simulations, there is evidence that desert dust is lifted to UTLS altitudes and is entrained into the ATAL (Fadnavis et al., 2013; Lau et al., 2018). Further, simulations show elevated aerosol concentrations of sulphate, nitrate, ammonium, black carbon and organic carbon within the ATAL with contributions of nitrate dominating (Gu et al., 2016; Fairlie et al., 2020). Moreover, Fairlie et al. (2020) found in model simulations for summer 2013 a different chemical compositions of the ATAL depending on the location within the Asian monsoon anticyclone with nitrate aerosol as the dominant component at the southern flank of the anticyclone.  
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Aircraft in situ measurements suggest that at lower altitudes the chemical composition of ATAL particles are dominated by carbonaceous and sulphate materials, consistent with the expectation that aerosol trends in the UTLS in the past decades are under increasing influence of sulphur emissions in Asia (Martinsson et al., 2014; Vernier et al., 2015; Fadnavis et al., 2019a). However, the first offline (balloon-borne filter samples) chemical analysis of ATAL particles suggested the presence of nitrate aerosol, but undetectable concentrations of sulphate ions (Vernier et al., 2018). Also, the evaluation of a variety of remote sensing measurements indicates a strong contribution of solid ammonium nitrate particles to the ATAL (Höpfner et al., 2019).

The deep convection in summer over the Indian subcontinent also causes lower concentrations of stratospheric tracers, in particular ozone, in the Asian monsoon anticyclone, whereas water vapour shows a local maximum in the monsoon anticyclone (e.g., Park et al., 2007; Bian et al., 2012; Randel et al., 2015; Brunamonti et al., 2018). The tropospheric signature inside the anticyclone at potential temperatures between  $\sim 360$  K and  $\sim 440$  K (corresponding to about 13 to 18 km) stems from convective transport typically up to the lower end of this range, followed by slow diabatic uplift superimposed on the anticyclonic motion, which Vogel et al. (2019) referred to as “upward spiralling range”. Brunamonti et al. (2018) denote the altitude range between maximum convective outflow and the cold point tropopause as the Asian tropopause transition layer (ATTTL). Above, the ATTTL, based on H<sub>2</sub>O measurements and trajectory calculations, Brunamonti et al. (2018) found a layer of a confined air mass in the lower stratosphere, which is consistent with the concept of an upward spiralling range. Pan et al. (2014) identified the tropical tropopause transition layer (TTL) using chemical tracer-tracer relationships in the tropics and over the Asian monsoon. Their comparison shows that the tracer-identified transition layer over the Asian monsoon is similar to the TTL, although the ATTTL is located at higher potential temperature levels. However, during the monsoon season the vertical upward transport caused by radiative heating at the top the Asian monsoon anticyclone is faster than elsewhere in the tropics (Vogel et al., 2019).

Particularly strong emissions of NO<sub>x</sub>, SO<sub>2</sub>, NH<sub>3</sub>, and particulate matter occur in Northern India and China (van der A et al., 2017; van Damme et al., 2018; Zheng et al., 2018), which impact ozone and aerosol in the upper troposphere. Ozone and aerosol changes in the upper troposphere in turn have radiative impacts over the region (Roy et al., 2017; Fadnavis et al., 2018; Bian et al., 2020) (Roy et al., 2017; Fadnavis et al., 2018, 2019a; Bian et al., 2020). However, after the year 2013, because of the implementation of new emission control measures in China, China’s anthropogenic emission of some pollutants decreased substantially (e.g., SO<sub>2</sub>, NO<sub>x</sub>, CO) while emissions of NH<sub>3</sub> and nonmethane volatile organic compounds did not change much during 2010-2017 (Zheng et al., 2018).

The confinement of air masses of tropospheric origin in the monsoon anticyclone also affects the aerosol particles constituting the ATAL (Yu et al., 2017; Vernier et al., 2018; Höpfner et al., 2019). Particles in the ATAL originate from ground sources (both gas-phase precursors and aerosol particles), which are lifted to UTLS altitudes by convection. The formation mechanism of the aerosol particles constituting the ATAL is not well characterised, but there is evidence that formation of secondary aerosol in the upper troposphere within the Asian monsoon anticyclone at high relative humidity and low temperature is important (Vernier et al., 2015, 2018; Yu et al., 2017).

Only a few measurements of size distributions of particles in the ATAL are available, but measurements with an Early size-resolved measurements using a balloon-borne optical particle counter (OPC) in August 1999 showed high number concentrations (0.7–0.8 particles cm<sup>-3</sup>) of aerosol particles with radii of 0.15–0.6  $\mu$ m between about 130–70 hPa in the Asian summer

monsoon (Kim et al., 2003; Tobo et al., 2007). Recent measurements with an OPC (Deshler et al., 2003) from Hyderabad, India and ~~measurements~~ with a Printed Optical Particle Spectrometer (POPS) (Gao et al., 2016) from Kunming, China, ~~indicate that ATAL particles~~ (both in 2015) also confirm the presence of the ATAL (Yu et al., 2017; Vernier et al., 2018). The OPC measurements indicate that the concentration of ATAL particles is highest close to the cold point tropopause and decline  
5 ~~towards greater altitudes; concentrations (up to about 25 particles per  $\text{cm}^{-3}$ ) are dominated by particles with a size of  $\approx 0.1 \mu\text{m}$  (Yu et al., 2017; Vernier et al., 2018).~~ in the  $r > 0.094 \mu\text{m}$  channel, whereas particle number concentrations for the  $r > 0.15 \mu\text{m}$  channel, and the  $r > 0.30 \mu\text{m}$  channel are lower by a factor of 30 and 300, respectively (Vernier et al., 2018, Fig. 9).

Based on CALIOP measurements, the summertime aerosol optical depth over Asia associated with the ATAL ~~is~~ has increased from  $\approx 0.002$  to  $0.006$  (Vernier et al., 2015), leading to a reduction in insolation of about  $0.6$  between 1995 and 2013, resulting  
10 in a short-term regional forcing at the top of the atmosphere of  $-0.1 \text{ W/m}^2$  and a surface cooling of roughly  $0.5$ .

The impact of the ATAL is also modulated by El Niño. During El Niño, the ATAL is thicker and broader over the Indian region, resulting in a reduction of the solar flux and a surface cooling of about  $1$  over North India. An elevated ATAL over South Asia exacerbates the severity of Indian droughts (Fadnavis et al., 2019b). The radiative forcing – compensating about one third of the comparable radiative forcing associated with the global increase in  $\text{CO}_2$  (Vernier et al., 2015). The regional  
15 radiative forcing caused by the ATAL, differs for clear and total sky conditions; total sky calculations show less shortwave radiative forcing over the monsoon region because of cloudiness (Vernier et al., 2015). It is likely that, over Asia in the past  $\sim 20$  years, the altered radiative forcing has led to summertime reductions in surface temperature, although this effect is not quantified yet. However, the radiative forcing caused by the ATAL ~~can further be enhanced by~~ could be compared with the global aerosol forcing caused by moderate volcanic eruptions since 2000, which translates into a surface cooling of  $0.05$  to  
20  $0.12 \text{ K}$  (Ridley et al., 2014).

The co-occurrence of the ATAL with volcanic eruptions in the region further enhances the radiative forcing by the ATAL; Fairlie et al. (2014, Fig. 8), based on monthly accumulations of CALIPSO-CALIOP aerosol data between 14 and 40 km altitude, reported a top of the atmosphere radiative forcing locally over Asia and Europe during July 2011 in response to the Nabro volcanic eruption of about  $-0.8$  to  $-1.5 \text{ W/m}^2$ . Vernier et al. (2015) found a positive trend in summertime UTLS aerosol optical  
25 depth of about  $0.004$ . Therefore, periods of volcanic eruptions are removed in some ATAL studies; because the volcanic signal is much stronger than the ATAL signal itself, a volcanic signal will mask the ATAL (e.g. Thomason and Vernier, 2013; Vernier et al., 2015).

The impact of the ATAL is furthermore modulated by El Niño. During El Niño, the ATAL is thicker and broader over the Indian region, resulting in a regional reduction of insolation (total sky radiative forcing) of around  $-0.1$  reduction of the solar  
30 flux and a surface cooling of about  $1 \text{ W/m}^2$  over the years 1997–2015, compensating about one third of the radiative forcing associated with the global increase in  $\text{K}$  over North India. An elevated ATAL over South Asia exacerbates the severity of Indian droughts (Fadnavis et al., 2019b).

The chemical signature of air masses within the monsoon anticyclone (e.g. tropospheric pollutants, water vapour) is exported to the Northern Hemisphere during summer and fall through quasi-isentropic transport from low latitudes (e.g., Ploeger et al.,  
35 2013; Vogel et al., 2014, 2016, 2019; Spang et al., 2015; Garny and Randel, 2016; Müller et al., 2016; Fadnavis et al., 2018; Rolf

et al., 2018; Yan et al., 2019). Because of this export of air from the Asian monsoon circulation to the Northern Hemisphere, the ATAL particles contribute significantly ( $\sim 15\%$ ) to the Northern Hemisphere stratospheric column aerosol surface area on an annual basis (Yu et al., 2017).

~~Only limited information on the chemical composition of the ATAL particles is available from measurements so far. From simulations, there is evidence that desert dust is lifted to UTLS altitudes and entrained into the ATAL (Fadnavis et al., 2013; Lau et al., 2018). Aircraft in situ measurements suggest that at lower altitudes the chemical composition of ATAL particles are dominated by carbonaceous and sulphate materials, consistent with the expectation that aerosol trends in the UTLS in the past decades are under increasing influence of sulphur emissions in Asia (Martinsson et al., 2014; Vernier et al., 2015; Fadnavis et al., 2019a). However, the first offline (balloon-borne filter samples) chemical analysis of ATAL particles suggested the presence of nitrate aerosol, but undetectable concentrations of sulphate ions (Vernier et al., 2018). Also, the evaluation of a set of remote sensing measurements indicates a strong contribution of solid ammonium nitrate particles to the ATAL (Höpfner et al., 2019). Moreover, Fairlie et al. (2020) found in model simulations for summer 2013 a different chemical compositions of the ATAL depending on the location within the Asian monsoon anticyclone. They found that nitrate aerosol is a dominant component of the ATAL on the southern flank of the anticyclone.~~

The Asian monsoon in the UTLS does not constitute a stable, persistent unimodal anticyclonic circulation (as it sometimes appears in climatologies), but can be bimodal (with an Iranian and a Tibetan mode) and moreover shows a strong day-to-day variability (Zhang et al., 2002; Yan et al., 2011; Vogel et al., 2015; Nützel et al., 2016). There is also a substantial inter-annual variability of the monsoon circulation with impact on the concentrations of tracers confined in the monsoon circulation (Santee et al., 2017; Yuan et al., 2019). This variability of the monsoon circulation impacts on the variability of trace-gas (Luo et al., 2018) and aerosol distributions (Lau et al., 2018; Yuan et al., 2019).

The strong convective activity in the Asian summer monsoon region also impacts the temperature in the monsoon region and the formation of cirrus clouds. Enhanced convection is linked to cold anomalies in the subtropical lower stratosphere (Park et al., 2007; Randel et al., 2015). Further, the occurrence fractions of cirrus in the middle to upper (16–18 km) tropical tropopause layer (TTL) (e.g., Fueglistaler et al., 2009) exhibit a pronounced maximum over the Asian monsoon region, both in observations and in model simulations (Ueyama et al., 2018). Convection is likely the dominant driver of localised upper tropospheric  $\text{H}_2\text{O}$  and cloud maxima in the region (Park et al., 2007; Ueyama et al., 2018). Ice clouds in the tropical deep convective regions are important as they exert a considerable net warming effect (Hong et al., 2016)(Pandit et al., 2015; Hong et al., 2016).

To address fundamental open questions and uncertainties regarding the anticyclone and the ATAL, a number of balloon-borne campaigns have been conducted in summer in the Asian monsoon region employing the Compact Optical Backscatter Aerosol Detector (COBALD) instrument (Bian et al., 2012, 2020; Vernier et al., 2018; Brunamonti et al., 2018). Here we present an analysis of the COBALD measurements in Nainital in Northern India in August and November 2016. The signature of the ATAL is clearly visible in most of the soundings in August (but not in every single one), but we find a substantial day-to-day variability of the ATAL, both in backscatter intensity and in altitude range of the ATAL. A Lagrangian back-trajectory analysis is performed in order to identify the air mass origin in the model boundary layer and the transport pathways of air parcels contributing to the ATAL over Nainital in August 2016. The paper is organised as follows: Sect. 2 describes the soundings

in Nainital and the CLaMS trajectory calculations, Sect. 3 presents the results of aerosol backscatter measurements, Sect. 4 discusses detailed trajectory calculations for three selected days as well as the simulation results for all balloon flight from Nainital. Finally, our results are discussed in Sect. 5 and the conclusions are given in Sect. 6.

## 5 2 Methods

The findings presented here are based on a set of balloon measurements performed in Nainital, Uttarakhand, India (29.35°N, 79.46°E); Nainital is located at an elevation of 1820 m above sea level. The Aryabhata Research Institute of Observational Sciences (ARIES) provided the campaign base to perform the launches. These measurements were conducted in August and November 2016 and are described in detail by Brunamonti et al. (2018, 2019). A brief summary is given here.

### 10 2.1 Balloon-borne instrumentation

The aerosol and cirrus measurements analysed here are based on balloon-borne backscatter measurements by the Compact Optical Backscatter Aerosol Detector (COBALD). COBALD operates at optical wavelengths of 455 nm (blue visible) and 940 nm (infrared) and was developed at the ETH Zürich (e.g. Wienhold, 2008; Brabec et al., 2012). It was hosted by a standard RS41-SGP meteorological radiosonde and the DigiCORA MW41 sounding system (Vaisala, Finland). In addition, the  
15 balloon payload included an electrochemical concentration cell (ECC, manufacturer: EN-SCI, USA) (Komhyr, 1969; Komhyr et al., 1995) for measuring the ozone mixing ratio and a cryogenic frost-point hygrometer (CFH, EN-SCI, USA) (Vömel et al., 2007, 2016) for measuring the water vapour mixing ratio (Brunamonti et al., 2019; Jorge et al., 2020). We use the pressure measured by the RS41 as the main vertical coordinate for all instruments. The ATAL analysis is mainly based on the COBALD 455 nm measurements ~~-, in particular on the -~~(see Appendix B). The COBALD data are expressed as backscatter  
20 ratio (BSR), ~~which is defined as i.e., the ratio of the COBALD raw signal (from particulates and air molecules) over the pure molecular scattering (derived from the ambient molecular number density, using the total-to-molecular backscatter coefficient. This is calculated by dividing the total measured signal by its molecular contribution, which is computed from the atmospheric extinction according to Bucholtz (1995), and using the air density derived from the RS41~~ temperature and pressure ~~measured by RS41)~~measurements. The colour index (CI), defined as the 940-to-455 nm ratio of the aerosol component of the BSR, i.e.  
25  $CI = (BSR_{940} - 1)/(BSR_{455} - 1)$  is a useful indicator of particle size and can thus be employed to discriminate aerosol and cirrus measurements (Cirisan et al., 2014; Vernier et al., 2015; Brunamonti et al., 2018). The CI for all balloon flights over Nainital in August 2016 is shown in Fig. A1 in Appendix A. For the detection of aerosol particles, we consider the short wavelength channel (455 nm), which ~~is more sensitive to smaller particles than the long wavelength channel (940). It also~~ provides a smaller value for the maximum uncertainty (1% and 5% for the shortwave and longwave channel, respectively) and is thus  
30 less noisy ~~(Brabec et al., 2012) (see Appendix B for details)~~(Brabec et al., 2012, see Appendix B for details).

No.	Date	Pressure [hPa]	pot. Temperature [K]	$\overline{\text{BSR}}_{455}$	$\sigma$	note
NT001	02-08-16	141 – 82	365 – 405	1.067	0.008	
NT002	03-08-16	122 – 82	368 – 400	1.092	0.013	bottom Cirrus
NT003	05-08-16	115 – 78	374 – 415	1.067	0.010	
NT004	06-08-16	135 – 95	364 – 388	1.083	0.009	
NT005	08-08-16	156 – 82	362 – 408	1.071	0.005	
NT007	11-08-16	105 – 88	367 – 400	1.070	0.024	bottom Cirrus
NT009	12-08-16	140 – 92	363 – 385	—	—	Cirrus
NT011	15-08-16	140 – 92	366 – 389	1.023	0.009	no ATAL
NT015	17-08-16	150 – 75	363 – 422	1.076	0.014	
NT017	18-08-16	150 – 75	362 – 422	1.065	0.008	
NT018	19-08-16	128 – 73	365 – 420	1.073	0.010	bottom Cirrus
NT023	21-08-16	115 – 80	372 – 404	1.056	0.011	
NT025	23-08-16	120 – 85	369 – 398	1.054	0.007	
NT027	26-08-16	95 – 83	382 – 398	1.080	0.004	bottom Cirrus
NT029	30-08-16	115 – 74	370 – 418	1.059	0.006	
NT033	10-11-16	140 – 92	365 – 394	1.037	0.006	post-monsoon
NT034	11-11-16	140 – 92	359 – 401	1.034	0.003	post-monsoon

**Table 1.** Table of all night-time balloon launches analysed here. Listed are number, date, top and bottom of the ATAL noted in both pressure and potential temperature, the mean value of the backscatter ratio ( $\overline{\text{BSR}}_{455}$  excluding cirrus contributions) and its standard deviation ( $\sigma$ ) (see Sects. 2.2 and 3.1 for details). Top of the ATAL is defined as the pressure level, where either the  $\text{BSR}_{455}$  profile has the strongest gradient or where  $\text{BSR}_{455}$  merge to the November background. The bottom is defined using the same criteria, except for cases where ATAL is limited below by a cirrus cloud (3, 11, 19 and 26 August). On 12 August, the UTLS is filled by a 5 km thick cirrus cloud, hence the ATAL cannot be diagnosed. On 15 August 2016, the UTLS was largely cloud-free but without any indication of the ATAL. The November soundings are post-monsoon without the ATAL. For soundings without the ATAL a reference pressure (140–92 hPa) and potential temperature (calculated from the measured temperature and pressure) range is given. This reference range allows comparisons with the ATAL observations to be made (details see Sect. 4). Measurements where the ATAL is limited below by a cirrus cloud, where a thick cirrus cloud or no ATAL was detected in the UTLS as well as the post-monsoon flights are indicated in the last column of the table (under ‘note’) in contrast to the normal ATAL measurements.

## 2.2 Campaign description

From Nainital, at the foothills of the Himalayas, 15 successful night-time balloon launches were conducted during the peak of the monsoon season in August 2016 (Tab. 1). All balloon soundings with a COBALD instrument were launched at night-time between 23:00 Indian Standard Time (IST) (corresponding to Coordinated Universal Time (UTC) +6:30 h) and next day 03:00 IST. Due to the high sensitivity of its photodiode detector, COBALD can only be deployed during night-time. The frequency of soundings was adjusted depending on the meteorological conditions. Out of a total of 30 balloon soundings (Brunamonti et al., 2018), we present here 17 launches (15 in August, 2 in November), which were performed with the COBALD instrument (Tab. 1). Four launches were excluded in our analysis. The 16 August launch burst below 3 km altitude due to harsh weather conditions and on 28 August the data showed very low signal-to-noise ratio in the stratosphere, likely caused by optical contamination. The post processing of the COBALD data requires the normalisation of the raw signal to a reference value, which is typically based on the measurements from above 30 km altitude (i.e. above the stratospheric aerosol layer). Due to the low absolute BSR signal of the ATAL, a careful calibration of all profiles is critical here. Since no stratospheric data are available on 5 and 12 August due to early bursts (below 20 km altitude), for the sake of consistency, we ~~exclude~~ excluded these two profiles from our analysis.

## 2.3 Data analysis and processing

In order to identify the ATAL, it is necessary to discriminate ~~aerosol~~ between aerosol particles and cirrus clouds (ice particles) in the backscatter measurements. For this purpose, we used the ice saturation ( $S_{ice}$ ) from the CFH instrument and the CI from the COBALD measurements (Cirisan et al., 2014). We ~~reject layers with CI > 7.0~~ define clear-sky (i.e. cloud-free) conditions: only layers are used in the ATAL analysis where the criteria CI < 7, BSR<sub>940</sub> ≥ 2 and S<sub>ice</sub> > 70% as cirrus clouds are simultaneously fulfilled. All other measurements are rejected from the ATAL analysis. The 70% threshold for S<sub>ice</sub> is on purpose low enough to ensure, that no cirrus cloud measurements are misinterpreted as aerosol measurements (Vernier et al., 2015; Li et al., 2018; Brunamonti et al., 2018). Other sections of the profiles measured during the August soundings, which show substantially elevated values of BSR<sub>455</sub> ~~in the UTLS~~ between ~140 and 70 hPa (≈ 14–18 km), are classified as ATAL. As we will see below, the classification is considerably simplified by the fact that the COBALD profiles reveal clearly the fingerprints of the ATAL, i.e. top and bottom of the aerosol layer can be identified with reasonable ~~precision~~ accuracy. We quantify the elevated values of BSR<sub>455</sub> by comparing the August profiles with the mean of the November measurements, when there is no ATAL during post-monsoon (Brunamonti et al., 2018). The enhancement of BSR<sub>455</sub> for the ATAL remains below 1.12 (and the CI below 7). Conditions with cirrus clouds embedded within the ATAL can also be easily identified, as the cirrus clouds have 10–100 times larger BSR<sub>455</sub> ~~As they~~ than ATAL particles. As cirrus clouds completely mask these ATAL particles, ~~these they~~ cannot be detected and quantified under such conditions (see Section 3 below).

Following earlier work (Brabec et al., 2012; Brunamonti et al., 2018), we use here vertically binned data for the CFH and COBALD instrument. BSR<sub>455</sub>, BSR<sub>940</sub>, CI and  $S_{ice}$  are binned in pressure intervals of 1 hPa for pressure greater than 300 hPa and 0.5 hPa for pressure less than 300 hPa, which yields an improved signal to noise ratio. This binning corresponds to a vertical

resolution of approximately 25 m in the UTLS. All data were carefully quality checked and measurements showing evidence of anomalous instrumental behaviour were rejected, as described by Brunamonti et al. (2018). The processing of COBALD includes the rejection of ‘moon spikes’ that may arise due to the oscillatory motion of the payload 60 m below the balloon, when the detector happens to be pointing towards the moon. Moon spikes affect only a tiny fraction of the COBALD data and care is taken not to confuse them with thin cirrus clouds (details see Appendix C).

## 2.4 Trajectory calculations

For the model analysis, we employ the trajectory module of the three-dimensional Lagrangian chemistry transport model CLaMS (McKenna et al., 2002; Pommrich et al., 2014) driven by dynamic fields from the European Centre of Medium-Range Weather Forecasts (ECMWF) interim reanalysis (ERA-Interim) employing a horizontal resolution of  $1^\circ \times 1^\circ$  (Dee et al., 2011). In ERA-Interim changes are implemented to improve deep and mid-level convection compared to previous reanalysis data (Dee et al., 2011). However, small-scale rapid uplift in convective cores is not included. Therefore convection over Asia is most likely underestimated in ERA-Interim. Nevertheless, upward transport in larger convective systems such as tropical cyclones is well represented in CLaMS trajectory calculations driven by ERA-Interim as shown by Li et al. (2017, 2020) by comparison with brightness and cloud top temperature derived from satellite observations. CLaMS back trajectory calculations are very well suited to analyse the detailed transport pathway of an air parcel, although they consider only the advective transport, neglecting mixing processes entirely. Earlier, CLaMS trajectories were applied to a variety of problems such as polar chlorine chemistry as well as transport in the tropics, in particular in the region of the Asian monsoon and in tropical cyclones (e.g., Vogel et al., 2003; Ploeger et al., 2010, 2012; Vogel et al., 2014, 2019; Li et al., 2017, 2018, 2020). To analyse the transport pathways and the origin of air masses contributing to the ATAL in August 2016 over Nainital, CLaMS 40-day backward trajectory calculations were performed. As the starting point for backward trajectory calculation we use the pressure levels of the measurements recorded every second (i.e. no binned data). The same trajectory model set-up is used as in several previous publications (Vogel et al., 2014; Li et al., 2017, 2018, 2020). In the CLaMS model, potential temperature is used as the vertical coordinate when the pressure is less than about 300 hPa, (i.e. in the upper troposphere and in the stratosphere); when the pressure is greater than about 300 hPa (more accurately, for pressure  $p$  exceeding a reference level of  $p/p_{\text{surface}} = 0.3$ ) a pressure-based orography-following hybrid coordinate  $\zeta$  (in units of K) is used (Ploeger et al., 2010; Pommrich et al., 2014). When the back trajectory dives into the CLaMS model boundary layer, we consider the air mass origin to be reached. The model boundary layer is defined as the hybrid pressure/potential temperature coordinate  $\zeta < 120$  K (Pommrich et al., 2014; Vogel et al., 2019), which corresponds to a layer  $\approx 2\text{--}3$  km above the surface of the Earth considering orography.

In general, trajectory calculations have limitations due to trajectory dispersion depending on the trajectory length. However, the frequently employed trajectory length to study transport processes in the Asian monsoon region is ranging from a couple of weeks to a few months depending on the transport times from Earth’s surface to atmospheric altitudes (e.g. Chen et al., 2012; Bergman et al., 2013). The CLaMS backward calculations to analyse the balloon measurements in Nainital show that the shortest transport times from Earth’s boundary layer to ATAL altitudes ( $\sim 360\text{--}420$  K, see Tab. 1) is about 10-15 days (see Sect. 4). Only a few trajectories are shorter. However, most of the trajectories show longer transport times than two weeks to reach ATAL altitudes

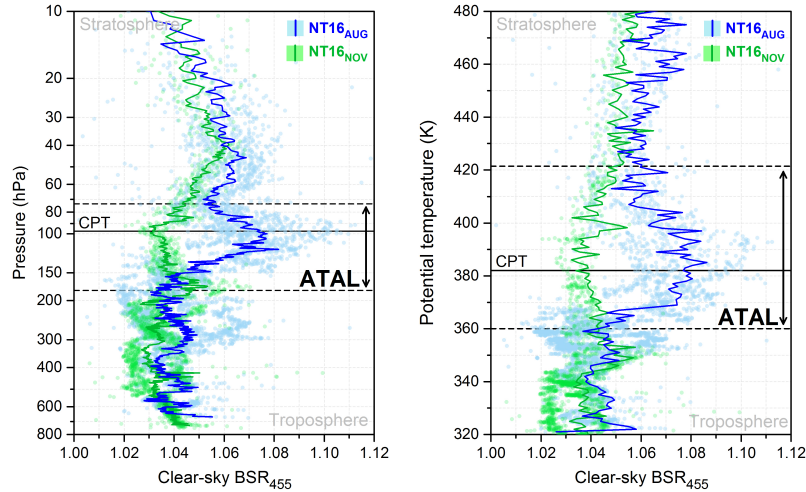


(up to  $\sim 420$  K or  $\sim 75$  hPa). The vertical upward transport of air parcels above the maximum level of convective outflow ( $\sim 360$  K) is determined by diabatic heating rates which are up to  $\sim 1\text{--}1.5$  K per day over the region of the Asian monsoon anticyclone during summer using ERA-Interim (Vogel et al., 2019). Convection occurs on short time scales of hours to a few days, whereby the transport from 360 K up to 420 K needs 40 to 60 days based on ERA-Interim. Therefore we performed

5 40-day backward trajectory calculations here, as well as trajectories with a length of 60 and 80 days to test the sensitivity on the trajectory length.

### 3 The observations in Nainital in 2016

#### 3.1 Aerosol backscatter measurements in August 2016



**Figure 1.** Backscatter measurements ( $\text{BSR}_{455}$ ) in Nainital, India, in August 2016 versus pressure (left) and versus potential temperature (right). Shown are  $\text{BSR}_{455}$  values for aerosol (dots) and averaged aerosol profiles (solid lines), both without cirrus contributions for August (blue) and November (green). Black lines show the mean temperature lapse-rate minimum (lower dashed), cold point tropopause (CPT, solid) and top of confinement (upper dashed). The black vertical arrows indicate the the ATAL region (Left panel: adapted from Fig. 11 of Brunamonti et al. (2018); Right panel: the ATAL boundaries and CPT from Tab. 3 in Brunamonti et al. (2018)).

An overview of the Nainital aerosol backscatter observations in August (blue) and November (green) 2016 with pressure as

10 the vertical coordinate is provided in Fig. 1 (left). Like for other years and locations (Vernier et al., 2015, 2018), the ATAL is clearly visible as an enhancement in backscatter ratio ( $\text{BSR}_{455}$ ) in August compared to the November measurements, with the averaged backscatter ( $\text{BSR}_{455}$ ) values in August reaching about 1.08 (Fig. 1). In Brunamonti et al. (2018), the ATAL is defined between the mean potential temperature lapse-rate minimum and the top of the confined lower stratosphere (Fig. 1,



dashed lines). The potential temperature lapse-rate minimum coincides approximately with the top of the convective outflow level ( $\approx 180$  hPa) (e.g., Gettelman and de Forster, 2002; Brunamonti et al., 2018).

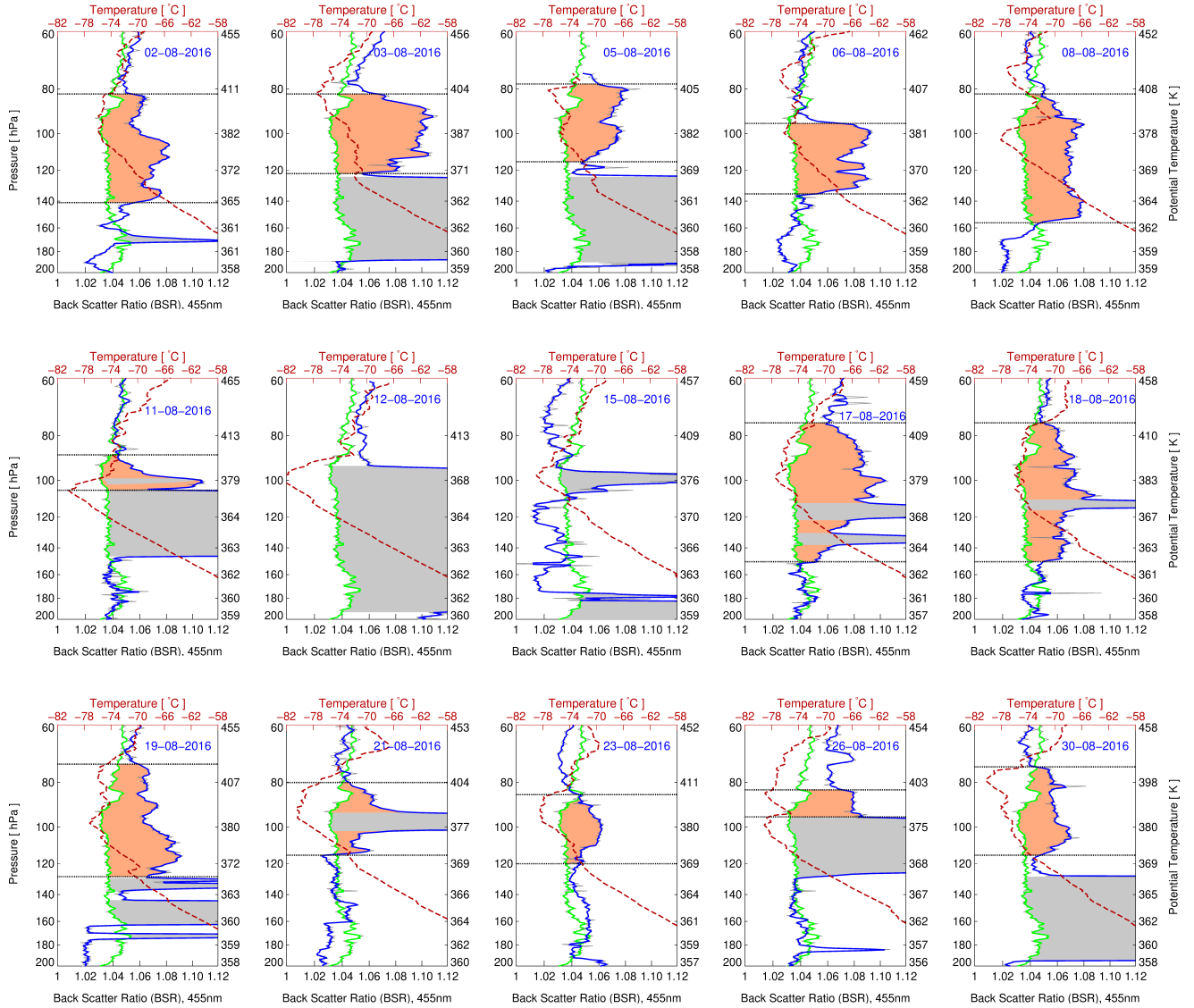
**Figure 1** (right) shows the same  $BSR_{455}$  values, but with potential temperature as the vertical coordinate. Using the backscatter measurements with potential temperature as the altitude scale, a somewhat different picture emerges than when using pressure as the altitude scale because the potential temperature has a large vertical gradient in this altitude region. In particular, a sharp increase in the averaged  $BSR_{455}$  in August around 360–365 K is clearly noticeable. This steep gradient represents the top of the convective outflow region (e.g., Gettelman and de Forster, 2002), i.e. below this level ~~more frequent deep convection scavenges the aerosols~~ aerosols are more frequently scavenged and removed by precipitation. An enhancement of the averaged  $BSR_{455}$  in August compared to November is noticeable up to potential temperatures of about 420 K ( $\sim 75$  hPa), but especially when considering potential temperature as the vertical coordinate, a top of the ATAL is not clearly defined. Only a slow decrease of averaged  $BSR_{455}$  with altitude is observed (Fig. 1, right). This observation is consistent with a decreasing confinement of the air mass within the Asian monsoon anticyclone with increasing potential temperature (Brunamonti et al., 2018; Vogel et al., 2019).

In Fig. 2, all 15 investigated COBALD balloon profiles in August 2016 (see also Tab. 1) are shown individually (blue lines) in comparison to the average of the two November measurements (green lines) used as a background signal. The top and bottom of the ATAL was determined for each balloon profile individually (quantified in Tab. 1) and these levels are shown in Fig. 2 as horizontal black dashed lines. Here we approximate the top and bottom of the ATAL by the pressure level where the  $BSR_{455}$  profile has the strongest gradients or where the  $BSR_{455}$  merges with the November background. The bottom is set using the same criteria as for the top of the ATAL, except for cases where a cirrus cloud was detected directly below the ATAL. In these cases the top of the cirrus cloud is set ~~to as~~ to as the bottom of the ATAL which is hidden by the cirrus cloud (3, 11, 19, 26 August).

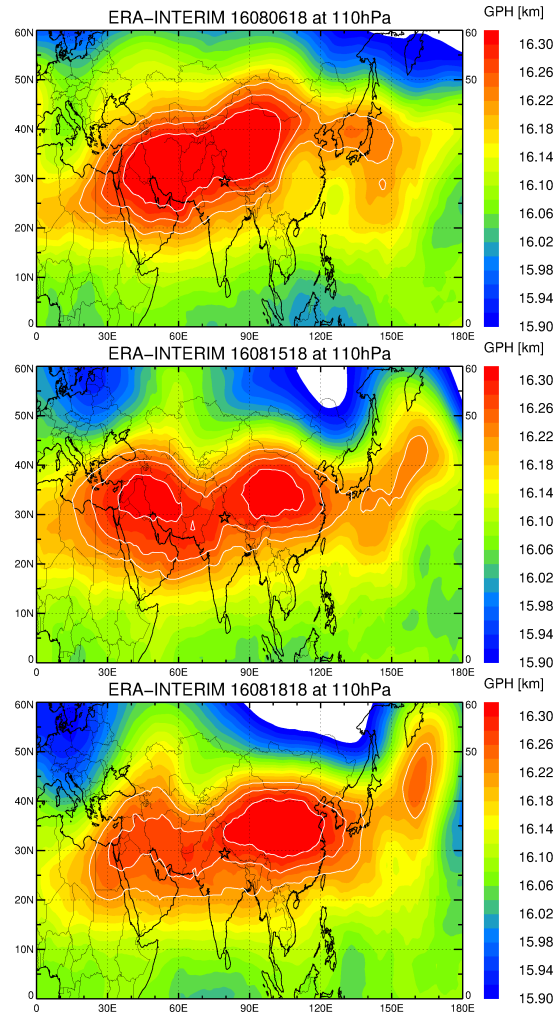
The mean value of the backscatter intensity ( $\overline{BSR}_{455}$ ) between bottom and top is individually calculated for each balloon sounding using the binned data (see Tab. 1). Cirrus clouds between top and bottom are excluded in the calculation of  $\overline{BSR}_{455}$  and its standard deviation. Therefore, no  $\overline{BSR}_{455}$  value is shown in Tab. 1 for the ‘no ATAL flight’ on 12 August, because over the entire altitude range where the ATAL is expected cirrus was detected.

### 3.2 Day-to-day variability of the ATAL

The enhancement of  $BSR_{455}$  for the individual measurements in August 2016 compared to the mean of the two November measurements is shown in Fig. 2 (orange shading). There is a strong variability of the altitude, the vertical extent and the  $BSR_{455}$  intensity within the ATAL. There are even days, when no ATAL is observed over Nainital (12 and 15 August 2016). In addition, on many balloon flights, a cirrus cloud is observed (e.g. as a very strong enhancement of  $BSR_{455}$ , see also section 2.3 above); cirrus clouds are marked in grey in Fig. 2. These cirrus clouds can occur below the ATAL, but there are also cases when cirrus clouds (ice particles) of a moderate vertical extent ( $\sim 10$  K) are measured within the ATAL (17, 18, and 21 August 2016). From the measurements alone we can not exclude that the ATAL and cirrus clouds can coexist in this region (see Sect. 2.3 and discussion in Sect. 5).



**Figure 2.** COBALD  $BSR_{455}$  measurements in the UTLS region versus pressure 60–200 hPa for each flight in August 2016 (blue line). Corresponding potential temperature calculated using both measured pressure and temperature is also given for orientation (see right-hand y-axis). The averaged  $BSR_{455}$  profile of the two November measurements is shown in green. The blue and the green lines show a slightly smoothed representation (three point smoothing) of the original 0.5 hPa binned data (light grey lines; see Sect. 2.3 for details). The measured temperature for each day in August 2016 is shown in red. The ATAL is highlighted by the orange shading and cirrus clouds are indicated by the grey shading. The top and bottom boundaries of the ATAL (see Tab. 1) are marked by horizontal dashed black lines, except for 12 and 15 August, as no ATAL was detected on these days.



**Figure 3.** Location of the Asian monsoon anticyclone measured by the geopotential height at 110hPa for 6 August, 15 August and 18 August 2016 at 06:18:00 UTC (corresponding to 02:23:30 local time (IST) in Nainital). The GPH of 16.22, 16.26, and 16.30 km are shown as white contour lines. The location of Nainital is marked by a black star symbol.

From a seasonal average perspective, the ATAL extends in the longitudinal direction from the Middle East to East Asia and meridionally between  $\sim 15^{\circ}$ - $45^{\circ}$ N (e.g., Vernier et al., 2015; Fairlie et al., 2020). This climatological picture is also true for the Asian monsoon anticyclone itself (e.g., Park et al., 2008; Vogel et al., 2016). Likewise, for the Asian monsoon anticyclone, there is a large variability from day to day in spatial extent, strength, and location, manifesting in an oscillation between a state with one anticyclone and two separated anticyclones (two modes) (e.g., Zhang et al., 2002; Yan et al., 2011; Vogel et al., 2015; Nützel et al., 2016).

In the following, we consider three specific days more closely, which are examples for particular cases (see also Sec. 4). We consider the case of an established ATAL (6 August 2016), a case, where no ATAL was observed (15 August 2016) and a case influenced by a typhoon (18 August 2016). We selected the 6 August because the mean value of the backscatter intensity ( $\overline{\text{BSR}}_{455}=1.083$ ) is the highest from all ATAL cases and because the measurement is not influenced by cirrus. From the two no ATAL cases (12 and 15 August), 15 August is chosen because the cirrus cloud is thinner than on 12 August.

The measurement in Nainital on 18 August 2016 was partially influenced by air uplifted in a tropical typhoon (this case will be discussed in detail in Sec. 4.3). On 18 August the ATAL occurs ~~in-an~~ over a broader altitude range compared to 6 August, however the intensity of the ATAL is ~~in-general~~ lower ( $\overline{\text{BSR}}_{455}=1.065$ ) than on 6 August.

The location of the Asian monsoon anticyclone for these three days is shown in Fig. 3. The Asian monsoon anticyclone is indicated here by geopotential height at 110 hPa ( $\text{GPH} = \Phi g^{-1}$  with geopotential  $\Phi [\text{m}^2 \text{s}^{-2}]$  and  $g = 9.81 [\text{m s}^{-2}]$ ) using ERA-Interim reanalyses for 6, 15, and 18 August 2016. The 110 hPa pressure level is selected because this level is located within the ATAL for most balloon flights in August 2016.

For the case of an established ATAL (6 August 2016) a single-modal anticyclone is found. For the case with no observation of an ATAL (15 August 2016), a bi-modal structure of the anticyclone is found and Nainital is located between the eastern and western mode of the Asian monsoon anticyclone. For the ATAL case impacted by a tropical cyclone (18 August 2016) a single-modal anticyclone is found with pronounced eddy shedding events (outflow from the anticyclone) towards both the east and the west (e.g. Vogel et al., 2015, 2016).

## 25 4 Results of the back-trajectory analysis

As shown in Fig. 2, there is a strong day-to-day variability in the ATAL altitude range, vertical extent and the  $\text{BSR}_{455}$  intensity. To analyse the transport pathway and the origin of the air masses contributing to the ATAL, for each sounding in August 2016 backward trajectories were calculated over a time period of 40 days. The dependence of the results of the trajectory calculations on the trajectory length (40, 60, and 80 days) will be discussed in detail below (Sec. 4.4).

30 The backward trajectories were started every second along the measured balloon profile where ~~an ATAL was~~ ATAL particles were detected during the flight (see Tab. 1 and Fig. 2, orange). The back-trajectory calculations are based on 1-sec data having a higher vertical resolution than the  $\text{BSR}_{455}$  values in Fig. 2 (binned data in pressure intervals of 0.5 hPa). In cases with a thin cirrus cloud embedded in the ATAL (17, 18, and 21 August 2016), the region of the cirrus cloud is included in the backward

No.	Date	BL %	LT %	UT %	LS %	Num. Traj.
		$\zeta \leq 120 \text{ K}$	$\zeta > 120 \text{ K}$ $\theta \leq 340 \text{ K}$	$\zeta > 120 \text{ K}$ $340 \text{ K} < \theta \leq 370 \text{ K}$	$\zeta > 120 \text{ K}$ $\theta > 370 \text{ K}$	
NT001	02-08-16	47.0	10.4	30.3	12.3	670
NT002	03-08-16	39.1	17.2	36.5	7.2	419
NT003	05-08-16	22.3	13.8	44.6	19.4	413
NT004	<b>06-08-16</b>	64.2	14.0	21.6	0.3	385
NT005	08-06-16	54.3	15.4	30.3	0.0	680
NT007	11-08-16	40.9	11.2	24.9	23.0	269
NT009	12-08-16	28.7	11.0	54.6	5.6	463
NT011	<b>15-08-16</b>	44.8	19.8	30.2	5.2	444
NT015	17-08-16	31.8	11.1	41.6	15.5	651
NT017	<b>18-08-16</b>	45.1	10.6	29.5	14.8	705
NT018	19-08-16	27.1	10.5	29.9	32.5	569
NT023	21-08-16	29.6	19.6	24.8	26.0	250
NT025	23-08-16	63.7	11.5	18.7	6.0	331
NT027	26-08-16	13.3	16.7	50.8	19.2	120
NT029	30-08-16	29.6	17.3	34.7	18.4	392
NT033	10-11-16	46.1	35.9	17.5	0.6	538
NT034	11-11-16	13.9	22.4	57.8	6.0	519

**Table 2.** Contributions of the origin of the air masses found in the ATAL using 40-day backward trajectories according to sources in the boundary layer (BL), in the lower troposphere (LT), in upper troposphere (UT) and in the lower stratosphere (LS). The number of backward trajectories (starting within the ATAL layer) calculated along each balloon profile is given in the last column. In the second and third row, the selection criteria for BL, LT, UT, and LS are listed. Trajectories are considered ending in the BL, when they are located for the first time below about 2–3 km (i.e., hybrid coordinate  $\zeta < 120 \text{ K}$ ). The location of this point is referred to as ‘end point’ of the trajectory in the model boundary layer. For the remaining trajectories ending at atmospheric altitudes ( $\zeta > 120 \text{ K}$ ), a potential temperature criterion ( $\theta$ ) is employed to discriminate between origins in LT, UT and LS. The dates of three balloon flights representing typical situations encountered during the measurements in August 2016 are marked in bold type: the case of an established ATAL (6 August 2016), a case without the observation of an ATAL (15 August 2016), and a case, where the measurements are heavily influenced by air masses uplifted in a tropical cyclone (18 August 2016).

trajectory calculation because it is likely that ATAL and ice particles coexisted in this region (see Sect. 2.3 and discussion in Sect. 5). For the two no ATAL cases (12 and 15 August 2016) as well as ~~both the two~~ post-monsoon measurements (10 and 11 November 2016), backward trajectories were started in a pressure range between 140 and 92 hPa, to allow a comparison with the ATAL observations.

- 5 We classify the origin of the air masses found in the ATAL according to sources in the model boundary layer, in the lower troposphere (LT), in the upper troposphere (UT) and in the lower stratosphere (LS) (Tab. 2). Trajectories are considered ending in the model boundary layer (BL) when they are located for the first time below about 2–3 km (i.e., hybrid coordinate  $\zeta \leq 120$  K). The location of this point is referred to as ‘end point’ of the trajectory in the model boundary layer. The trajectory length for these trajectories ending in the BL is shorter than 40 days because they reach the BL before 40 days. Within 40 days  
 10  $\approx 14$ –64% of the trajectories are reaching the model boundary layer. For the remaining trajectories ending at atmospheric altitudes, a potential temperature criterion is employed to discriminate between origins in the lower troposphere (LT), in the upper troposphere (UT) and in the lower stratosphere (LS) (see Tab. 2).

- There is a strong variability of the contributions of different atmospheric ~~height-altitude~~ layers (BL, LT, UT and LS) to the air masses in the ATAL (Tab. 2), demonstrating that within the ATAL a mixture of air masses of different origin ~~existexists~~.  
 15 Before we discuss the results of the back-trajectory calculations for all balloon-flights in Nainital, three days representing typical situations encountered during the measurements in August 2016 are presented in detail: the case of an established ATAL (6 August 2016), a case without the observation of an ATAL (15 August 2016), and a case, where the measurements are heavily influenced by air masses uplifted in a tropical cyclone (18 August 2016).

#### 4.1 Case 1: Established ATAL on 6 August 2016

- 20 On 6 August 2016 the ATAL was observed between 364 K to 388 K (135–95 hPa). The back-trajectory analysis of 40 days (Tab. 2) shows that 64% of the trajectories are from the boundary layer (BL), 14 % from the lower troposphere (LT), 22 % from the upper troposphere (UT) and only 0.3 % from the lower stratosphere (LS). The 40-day backward trajectories (potential temperature versus time) classified by the location of the trajectory end points at different atmospheric ~~height-altitude~~ layers are shown in Fig. 4.

- 25 Air masses from the BL are mainly uplifted very fast by individual ~~convection-convective~~ events within short time periods of 1–2 days up to  $\approx 360$  K. Subsequently, slower updraft above 360 K occurs within the Asian monsoon anticyclone (at longitudes between 0°E and 140°E) where the air masses follow the anticyclonic flow (see Fig. 5). This region is referred to ‘upward spiralling range’ by Vogel et al. (2019). Trajectories from the LT are very similar to those from the BL, however there are more trajectories originating outside of the Asian monsoon region from western longitudes. Most trajectories from the  
 30 UT experienced radiatively driven updraft within the upward spiralling range. In contrast, the trajectory originating in the LS descends in the stratosphere (coming from Northern America) and subsequently ~~aseent-occurs~~ ascends within the anticyclone.

The trajectories originating from the BL are from the region of the Asian monsoon anticyclone and from the western Pacific (Fig. 5). To obtain a deeper insight into the origin of air masses contributing to the ATAL, trajectories originating in the BL are further separated. In a latitude-longitude box from 60°E to 160°E and from 5°S to 45°N (Fig. 6, Tab. 3), we distinguish

between Tibetan Plateau (Tibet), foothills (Foothills) including the Himalayan foothills, the remaining continental (Land) and maritime area (Ocean). The continental areas were separated according to geopotential ( $\Phi$ ) or GPH ( $\Phi g^{-1}$ ). The Tibetan Plateau is defined as the altitude above a geopotential  $\Phi$  of 40000 [ $m^2 s^{-2}$ ] (above GPH  $\approx 4000$  m), the foothills as  $20000 < \Phi < 40000$  [ $m^2 s^{-2}$ ] (GPH  $\approx 2000$ –4000 m), the remaining continental area as  $\Phi < 20000$  [ $m^2 s^{-2}$ ] (below GPH  $\approx 2000$  m).

5 Trajectories originating outside the latitude-longitude box are classified as Residual.

~~Fig~~Figure. 6a shows the location of the end points of all trajectories ending in the BL for 6 August 2016 colour-coded by the transport time from the boundary layer to the location of the measurements. The shortest transport times of about 15–20 days are found for trajectories originating on the Tibetan Plateau. In addition there is a cluster of trajectories from northeast India with transport times of about 25 days.

10 In addition, the location of the maximum updraft within 18 hours along these 40-day backward trajectories is calculated. It is first determined where the maximum change in potential temperature occurs ( $\Delta\Theta_{\max}$ ) along the 40-day backward trajectories (calculated as running mean over the change in potential temperature within 18 hours). The location of the maximum updraft within ~~the~~these 18 hours is then calculated as the mean location of the trajectory within 18 hours for  $\Delta\Theta_{\max}$  (Fig. 6b).

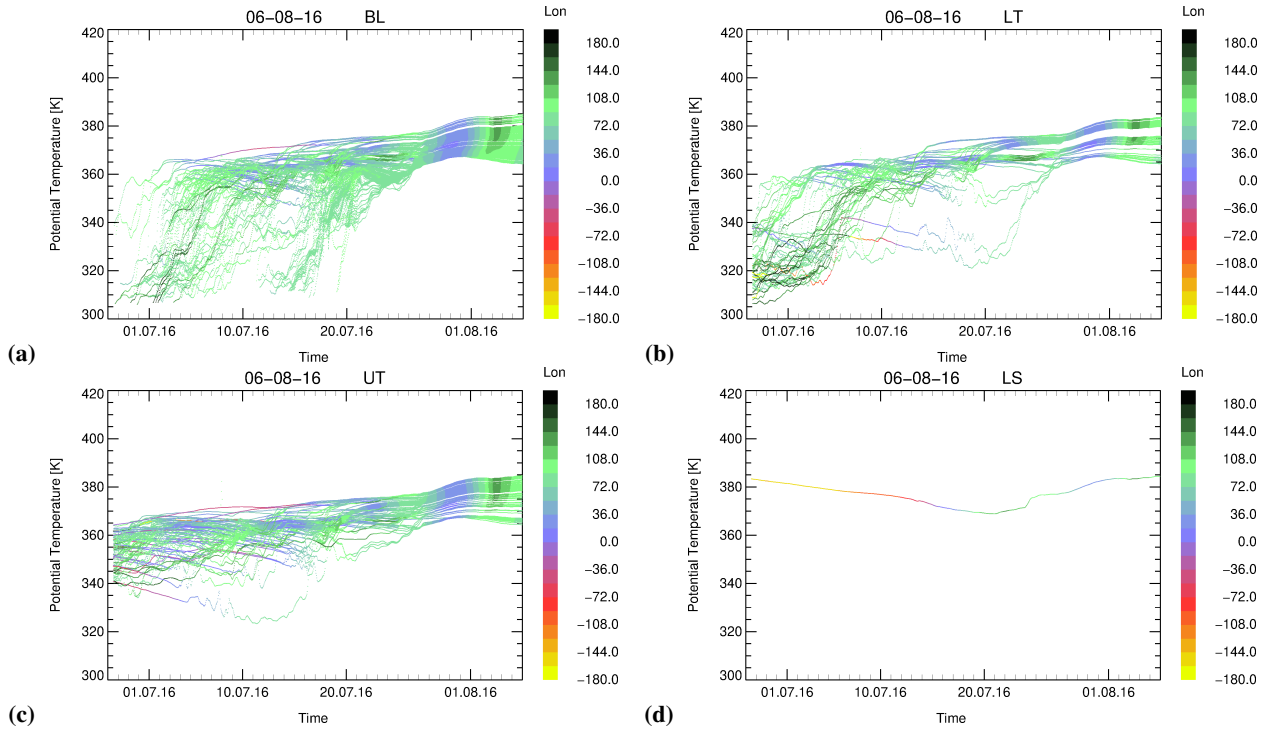
It is evident that the location of the end points (Fig. 6a) and of the strongest updraft (Fig. 6b) differ substantially. Our  
 15 calculations show a cluster of locations with an updraft larger then  $25 K 18h^{-1}$  within a belt along the southern edge of the Himalayas and over Myanmar. The area of the foothills (GPH  $\approx 2000$ –4000 m) is very small because the gradient of the GPH is very steep at the southern slope of the Himalayas, therefore it is difficult to highlight this belt using only the GPH as criterion to discriminate the foothills. ~~Nevertheless, the~~The number of trajectories with the location of strongest updraft on the ~~foothills~~  
~~(9%) or on the~~ Tibetan Plateau (~~36~~44%) is ~~slightly~~ enhanced compared to the end points of these trajectories (~~8%, 34%~~) (Tab. 3  
 20 and Tab. 4).

The location of  $\Delta\Theta_{\max}$  depends strongly on the time interval over which the change in potential temperature is calculated. If a very short time interval is chosen, the deduced region of strongest updraft depends only on one input data set at one particular time step. However, the longer the time interval the higher the uncertainty of the location of maximum updraft along the trajectories. This is because the vertical updraft by convection in CLaMS trajectories is driven by vertical velocities  
 25 deduced from ERA-Interim and is not restricted to very narrow regions (e.g. see Fig. 4). Therefore, the locations of  $\Delta\Theta_{\max}$  are spread out increasingly over the region of the southern slope of the Himalayas with an increasing time interval. To avoid the uncertainty caused by a larger time interval and because of the fact that convection occurs on a time ~~frame~~scale of a few hours, we decided to use a time interval of 18 hours. This choice also ensures that at least 3 input data sets from ERA-Interim (with 6 hour resolution) are included to localise the preferred regions of strongest updraft in the region of the Asian monsoon.

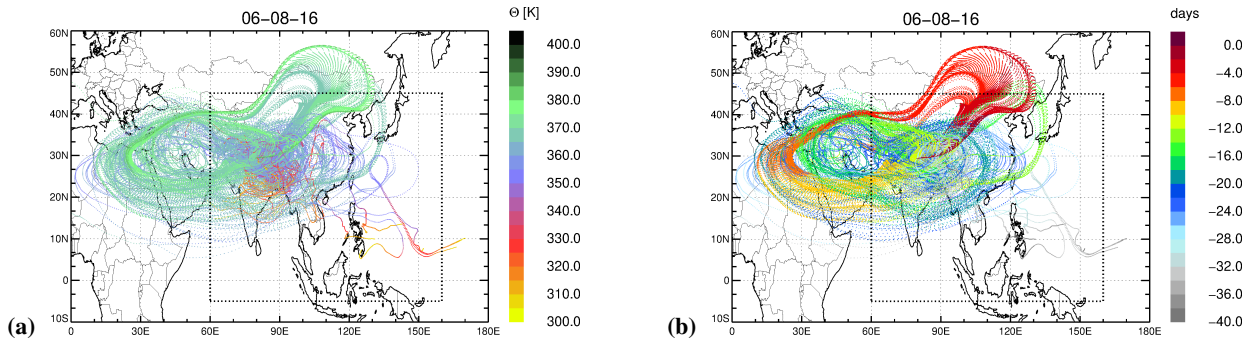
## 30 4.2 Case 2: No ATAL on 15 August

On 15 August 2016 no ATAL was detected by the COBALD measurements. To compare this flight with the other cases where an ATAL was probed during August 2016, CLaMS backward trajectories are initialised in a pressure range between 140 hPa to 92 hPa (365 K to 389 K). In this pressure range 45% of the trajectories are from the BL, that is 20% less than the BL contribution of Case 1 (64%). Therefore, much higher contributions of the UT (30%) and LT (20%) as well as from the LS (5%) are found



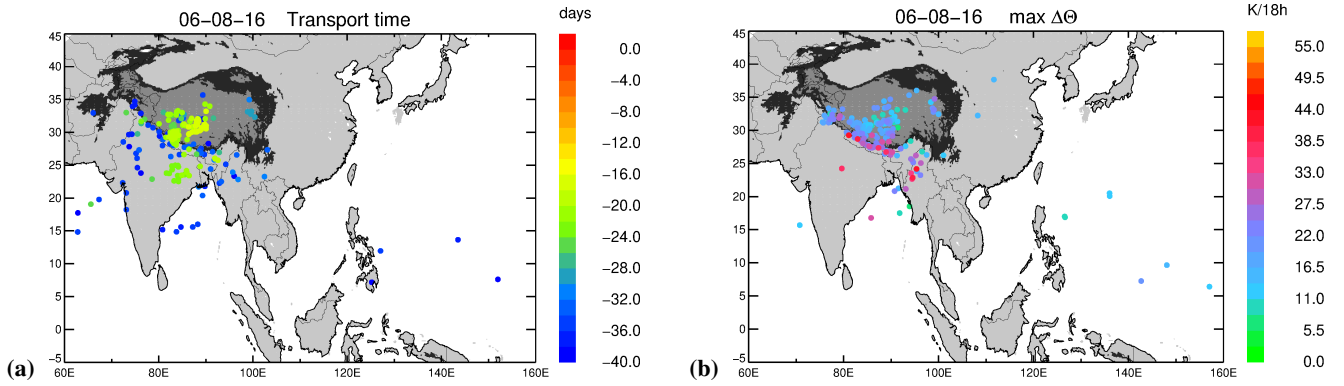


**Figure 4.** 40-day backward trajectories initialised on 6 August 2016 at the location of the ATAL measurement (see Tab. 1). The trajectories are sorted by air mass origin in the boundary layer (BL), in the lower troposphere (LT), in the upper troposphere (UT) and in the lower stratosphere (LS). The fractions of the different atmospheric height-altitude layers (BL, LT, UT, LS) to the composition of the ATAL are shown in Tab. 2. The colour indicates the location of the trajectory in longitude.



**Figure 5.** 40-day backward trajectories initialised within the ATAL on 6 August 2016 originating in the BL (same trajectories as shown in Fig. 4a) colour-coded by potential temperature (a) and by days back from 6 August 2016 (b). In a latitude-longitude box from 60°E to 160°E and from 5°S to 45°N indicated by dashed lines the air mass origin in the BL is further analysed (see Fig. 6, Tab. 3)





**Figure 6.** (a) Location of end points of the trajectories, initialised on 6 August within the ATAL, where they reach the model boundary layer (BL) within 40 days. The end points are colour-coded by the transport time between BL and measurement. (b) Location of the strongest updraft along the 40-day backward trajectories colour-coded by the strongest change in potential temperature within 18 hours [in  $\text{K } 18\text{h}^{-1}$ ]. The Tibetan Plateau (dark grey), foothills (black), the remaining continental area (Land; light grey) and maritime area (Ocean; white) are indicated (see Tab. 3).

on 15 August in contrast to Case 1 (22%, 14%, 0.3%) (see Tab. 2). The lesser contribution of the BL can be explained by the location of the anticyclone related to the location of the measurements. On 6 August 2016 the Asian monsoon anticyclone was over Nainital, while on 15 August Nainital was located between the western and the eastern mode of the anticyclone (see Fig. 3).

- 5 The 40-day backward trajectories for the 15 August 2016 are shown in Fig. 7. In contrast to Case 1 no pronounced individual convection events are noticeable in the trajectories from BL and LT on 15 August. Here, an alternating up and downward transport is found along most of the trajectories below 360 K and only a few trajectories show a strong upward transport (convection) up to 360 K within 1–2 days.

- 10 ~~Depending on the altitude of the measurements two branches~~ Focusing on the trajectories from the BL (Fig. 8) two branches,  
a western and eastern branch, of trajectories are found depending on the altitude of the measurements (Fig. 7a, ~~b and c~~);  
~~the higher ones~~); trajectories from above 370 K are from eastern and ~~the lower ones~~ trajectories from below 370 K are from  
western longitudes. ~~Focusing on the trajectories from the BL (Fig. 8), in~~ In the western branch of the trajectories the air masses  
are transported around the western mode of Asian monsoon anticyclone, while air masses from the eastern branch are coming  
from the Pacific ocean and are transported along the southern edge of the eastern mode of the anticyclone to the measurement  
15 location in Nainital. During the first half of August several tropical storms occurred in the western Pacific. Typhoon Omais was  
active between 2–12 August 2016 (<https://www.jma.go.jp/jma/jma-eng/jma-center/rsmc-hp-pub-eg/besttrack.html>; storm ID:  
1605 (last access: 26 May 2020) and [https://en.wikipedia.org/wiki/2016\\_Pacific\\_typhoon\\_season](https://en.wikipedia.org/wiki/2016_Pacific_typhoon_season) (last access: 26 May 2020))  
and impacted the sounding on 15 August (see Fig. 9).

No.	Date	BL %	Tibet %	Foothills %	Land %	Ocean %	Residual %
NT001	02-08-16	47.0	18.5	3.0	16.9	6.9	1.8
NT002	03-08-16	39.1	13.8	2.4	8.4	9.1	5.5
NT003	05-08-16	22.3	7.5	1.7	8.0	4.1	1.0
NT004	<b>06-08-16</b>	64.2	33.5	7.5	17.7	4.2	1.3
NT005	08-08-16	54.3	22.6	3.8	21.9	4.6	1.3
NT007	11-08-16	40.9	22.3	2.6	14.1	1.5	0.4
NT009	12-08-16	28.7	14.3	1.1	7.6	4.5	1.3
NT011	<b>15-08-16</b>	44.8	16.9	2.7	16.2	5.2	3.8
NT015	17-08-16	31.8	16.4	2.6	7.7	3.5	1.5
NT017	<b>18-08-16</b>	45.1	3.7	0.6	7.2	29.5	4.1
NT018	19-08-16	27.1	7.7	1.1	8.8	6.7	2.8
NT023	21-08-16	29.6	3.2	1.2	3.2	13.2	8.8
NT025	23-08-16	63.7	23.6	3.0	19.9	14.5	2.7
NT027	26-08-16	13.3	2.5	0.0	3.3	6.7	0.8
NT029	30-08-16	29.6	6.1	2.3	7.9	11.5	1.8
NT033	10-11-16	46.1	0.0	0.0	4.1	25.8	16.3
NT034	11-11-16	13.9	0.0	0.0	0.0	1.0	12.9

**Table 3.** Fraction of 40-day backward trajectories (for all soundings in August 2016), which are ending in the boundary layer (BL) separated into the Tibetan Plateau (Tibet), foothills (Foothills), the remaining continental area (Land) and the maritime area (Ocean) in a latitude-longitude box from 60°E to 160°E and from 5°S to 45°N shown in Fig. 6, and finally all remaining trajectories reaching the BL outside the latitude-longitude box (Residual). The fraction 100% - BL% are those trajectories, which are from the LT, UT and LS (i.e. do not reach the BL within 40 days as shown in Tab. 2). The Tibetan Plateau is defined as the altitude above a geopotential  $\Phi$  of 40000 m<sup>2</sup>s<sup>-2</sup> (about 4000 m), the foothills as 21000 <  $\Phi$  < 40000 m<sup>2</sup>s<sup>-2</sup> (about 2000–4000 m), the remaining continental area as  $\Phi$  < 21000 m<sup>2</sup>s<sup>-2</sup> (below about 2000 m).

**Fig-Figure** 9a shows the location of the end points of back-trajectories in the BL colour-coded by the transport time from the BL to the location of the measurement. Lesser contributions from the Tibetan Plateau (17%) and the foothills (3%) are found compared to Case 1 (34%, 8%). Due to the dynamics of the bi-modal anticyclone the locations of the end points in the BL are much more widely spread over Asia (e.g. Pakistan, Afghanistan, China), the western Pacific, and the residual **Earth's surface** **surface of the Earth** ( $\approx 5\%$ ; see Tab. 3) as compared to Case 1. In Case 1, the end points are more clustered demonstrating the more frequent occurrence of individual **convection-convective** events. A few trajectories ending on the Tibetan Plateau have short transport times of about 10 to 12 days; these are shorter times than found for Case 1 (Fig. 6) and are caused by convection between 2 and 5 August.

No.	Date	BL %	Tibet %	Foothills %	Land %	Ocean %	Residual %
NT001	02-08-16	47.0	<del>17.2</del> 20.4	<del>9.0</del> 6.6	<del>17.8</del> 17.3	<del>2.8</del> 2.7	<del>0.3</del> 0.0
NT002	03-08-16	39.1	<del>15.3</del> 18.6	<del>4.3</del> 3.1	<del>9.8</del> 7.6	9.3	0.5
NT003	05-08-16	22.3	<del>9.0</del> 9.7	<del>3.4</del> 3.1	<del>8.0</del> 7.5	1.7	0.2
NT004	<b>06-08-16</b>	64.2	<del>36.1</del> 43.6	<del>8.8</del> 2.9	<del>15.8</del> 14.3	3.4	0.0
NT005	08-08-16	54.3	<del>25.9</del> 29.7	<del>9.3</del> 7.8	<del>17.2</del> 15.1	<del>1.8</del> 1.6	<del>0.1</del> 0.0
NT007	11-08-16	40.9	<del>24.9</del> 26.4	<del>3.7</del> 2.6	<del>11.5</del> 11.2	0.7	0.0
NT009	12-08-16	28.7	<del>11.4</del> 15.3	<del>7.1</del> 3.5	<del>6.9</del> 6.7	3.0	0.2
NT011	<b>15-08-16</b>	44.8	<del>16.0</del> 16.4	5.2	<del>17.8</del> 17.3	4.3	1.6
NT015	17-08-16	31.8	<del>15.7</del> 17.2	<del>5.5</del> 4.9	<del>6.6</del> 5.7	3.7	0.3
NT017	<b>18-08-16</b>	45.1	<del>3.4</del> 3.8	<del>1.6</del> 0.9	<del>6.4</del> 6.2	<del>27.2</del> 33.2	<del>6.5</del> 1.0
NT018	19-08-16	27.1	<del>7.6</del> 9.3	<del>3.7</del> 2.5	8.8	<del>5.4</del> 5.3	<del>1.6</del> 1.2
NT023	21-08-16	29.6	3.2	0.8	<del>3.2</del> 2.8	<del>17.6</del> 18.4	<del>4.8</del> 4.4
NT025	23-08-16	63.7	<del>24.8</del> 26.0	7.9	<del>22.4</del> 23.0	<del>7.3</del> 6.0	<del>1.5</del> 0.9
NT027	26-08-16	13.3	3.3	0.0	1.7	8.3	0.0
NT029	30-08-16	29.6	<del>6.9</del> 6.6	3.1	<del>7.1</del> 7.9	<del>12.5</del> 12.0	0.0
NT033	10-11-16	46.1	0.0	0.0	2.6	<del>31.2</del> 31.8	<del>12.3</del> 11.7
NT034	11-11-16	13.9	0.0	0.0	0.2	0.8	12.9

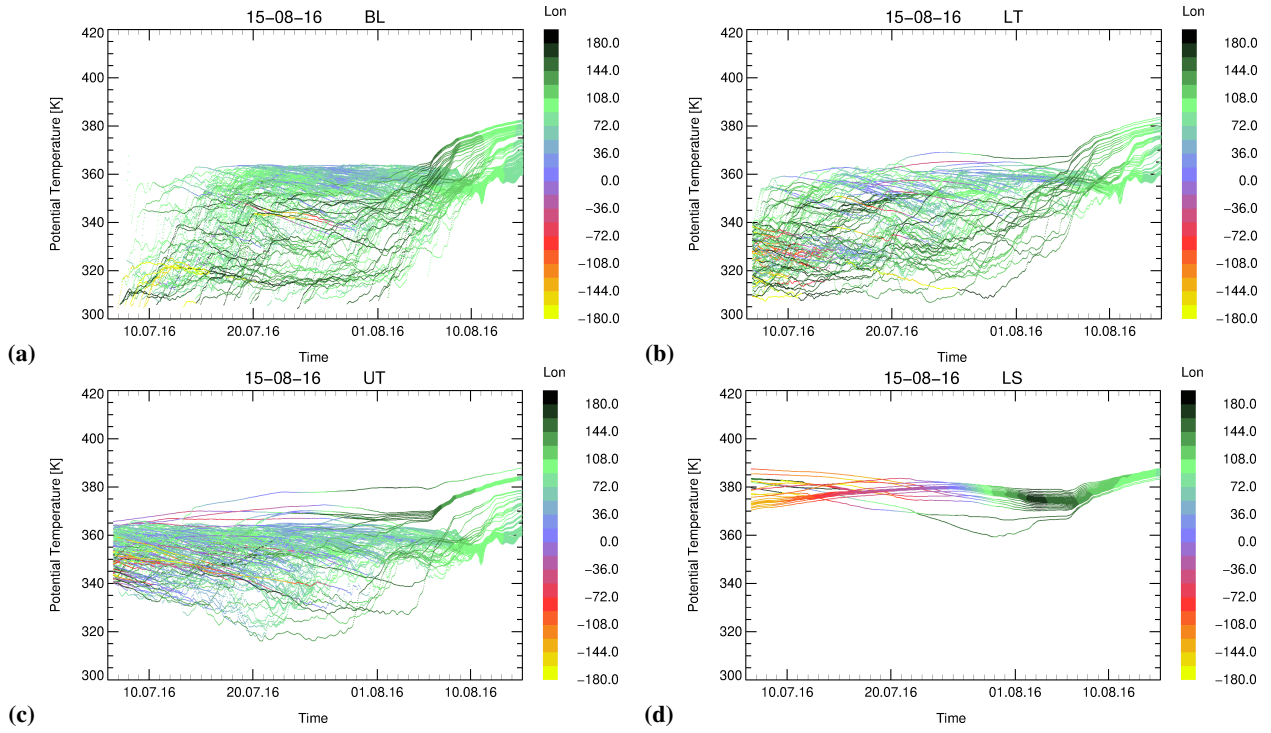
**Table 4.** Same as Tab. 3, but for the location of strongest updraft along the 40-day backward trajectories ending in the BL. The strongest updraft is calculated by the maximum change in potential temperature within 18 hours along the trajectories. The location of the maximum updraft within the 18 hours is then calculated as the mean location of the trajectory within 18 hours.

Similar as for Case 1, in Case 2 the location of the end points in the model BL (Fig. 9a) and of the strongest updraft (Fig. 9b) differ substantially. A cluster of locations with an updraft larger than  $25 \text{ K } 18\text{h}^{-1}$  is found at the southern edge of the Himalayas, over Myanmar, and ~~west~~east of the Tibetan Plateau.

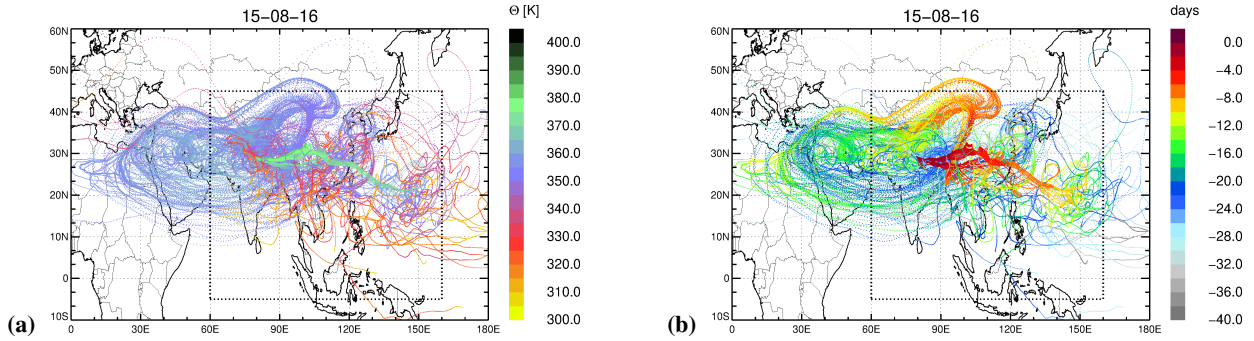
In Fig. 9, the location of the end points in the model BL of the strongest updraft for trajectories started in the cirrus cloud found between 376 and 381 K potential temperature on 15 August (see Fig. 2) are marked by a star symbol. End points in the BL of the cirrus cloud are found in different regions, demonstrating that the air masses in the cirrus cloud are a mixture of different origins. Thus the locations of strongest updraft for trajectories initialised within the cirrus cloud are found over the Pacific as well as over continental areas. Cirrus formation ~~as well as likely~~and the details of the transport pathways that lead to cirrus formation are not represented in our trajectory calculations based on ERA-Interim reanalysis.

#### 10 4.3 Case 3: Typhoon influence on 18 August

On 18 August the ATAL occurs over a broader potential temperature range from 362 K to 422 K compared to 6 August (364 K to 388 K, Case 1), however the the ATAL intensity ( $\overline{\text{BSR}}_{455}=1.065$ ) is lower compared to Case 1 ( $\overline{\text{BSR}}_{455}=1.083$ ) (Fig. 2 and

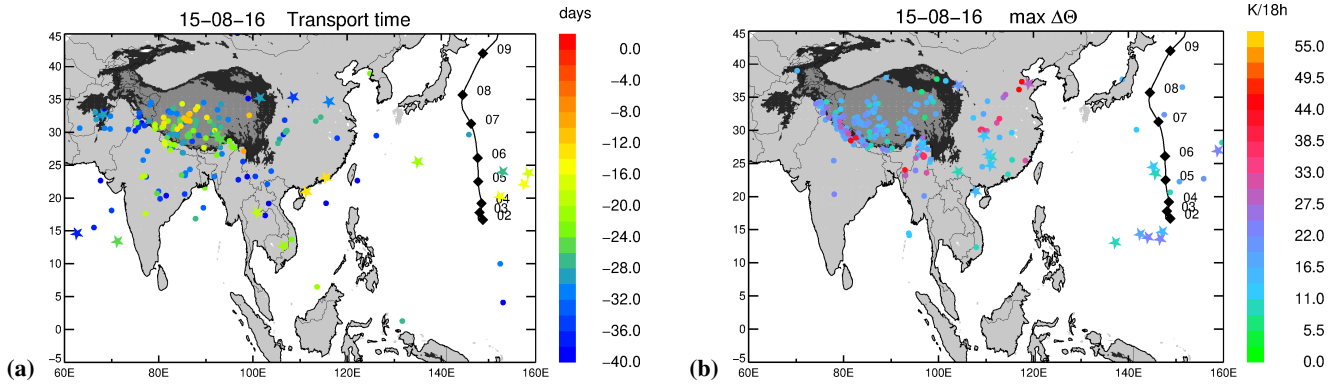


**Figure 7.** The same as [As](#) Fig. 4, but for Case 2 the no ATAL case on 15 August 2016. The trajectories are calculated in a pressure range between 140 hPa to 92 hPa (365 K to 389 K).



**Figure 8.** The same as [As](#) Fig. 5, but for the balloon flight on 15 August 2016 (Case 2). In addition to Case 1 shown in Fig. 5, the impact of tropical cyclone activity in the western Pacific is found in Case 2.

Tab. 1). The contributions from the BL are 45% and similar to Case 2, but much lower than for Case 1 (64%, Tab. 2). For Case 3, a stratospheric contribution of 15% is found, which is much higher than for Case 1 (0.3%) and Case 2 (5%) because of the considered top level of potential temperature (Case 1 up to 388 K, Case 2 up to 389 K and Case 3 up to 422 K) (see Figs. 4d, 7d



**Figure 9.** The same as [As Fig. 6](#), but for the no ATAL case on 15 August 2016. Locations of end points and locations of the strongest updraft of trajectories initialised within the cirrus cloud are indicated by a star symbol. The 40-day backward trajectories are impacted by typhoon Omais. Its storm track is shown (black line) and the location at noon time is additionally marked (black diamonds); the numbers indicate the day in August 2019.

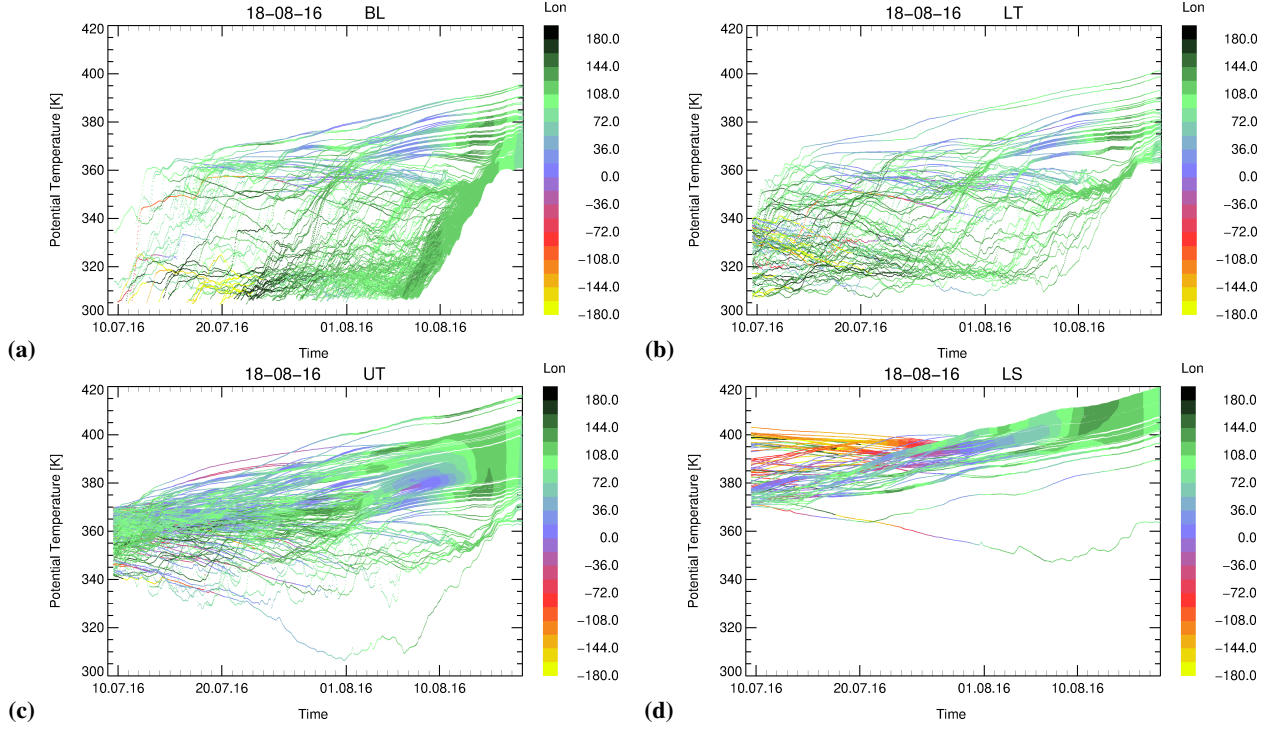
and 10d). The stratospheric contributions are air masses from the northern extra-tropical lower stratosphere. These air masses are transported along the subtropical jet and subsequently were slowly uplifted in the anticyclonic large-scale upward spiral around the Asian monsoon anticyclone by diabatic heating. In general, the higher within the upward spiralling range at the top of the Asian monsoon anticyclone, the more contributions from the stratospheric background are found (Vogel et al., 2019).

- 5 [Fig. Figure 10a](#) shows very strong convection between 6 and 15 August with strong updraft within a few days up to 360 K. These air masses originate from the western Pacific and [the Bay of Bengal](#) and are transported from the western Pacific directly to Nainital (Fig. 11). Another branch of BL air from the western Pacific is transported around the outer edge of the Asian monsoon anticyclone to Nainital (Fig. 11).

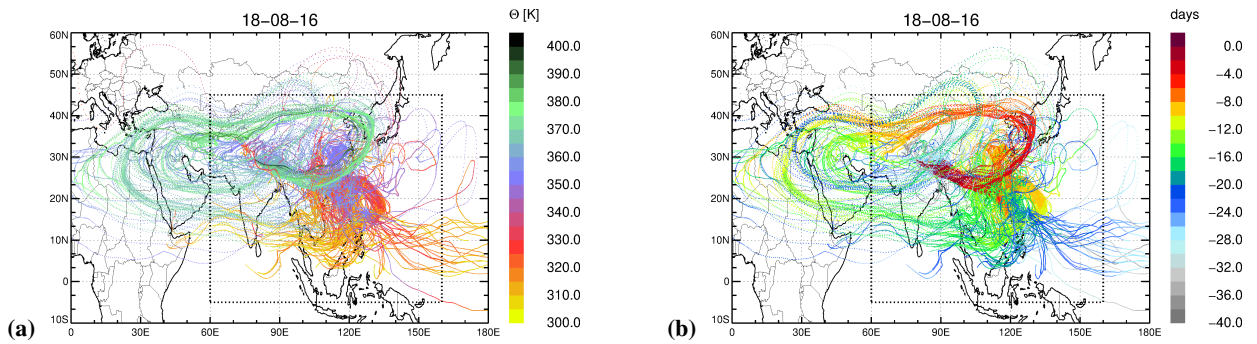
- 10 The location of the end points in the BL for Case 3 are shown in more detail in Fig. 12. The major fraction of the BL contribution is from the Ocean (30%) in contrast to Case 1 (4%) and Case 2 (5%). Also the location of  $\Delta\Theta_{\max}$  is found in the western Pacific, however at slightly different locations as the end points. The strong updraft over the Pacific is caused by tropical cyclone activity. The typhoon Nida was active between 29 July and 3 August over the western Pacific (<https://www.jma.go.jp/jma/jma-eng/jma-center/rsmc-hp-pub-eg/besttrack.html>; storm ID: 1604 (last access: 26 May 2020) and [https://en.wikipedia.org/wiki/2016\\_Pacific\\_typhoon\\_season](https://en.wikipedia.org/wiki/2016_Pacific_typhoon_season) (last access: 26 May 2020)) and impacted the balloon sounding on 18 August 2016.
- 15 Thus, in Case 3 polluted air masses within the ATAL [layer](#) measured below 400 K are diluted by air from the maritime boundary layer. At higher potential temperature levels (above 400 K) mixing with air masses from the stratosphere occurs.

- In Fig. 12, the location of the end points in the model BL of the strongest updraft for trajectories started in the cirrus cloud found between 370 and 373 K potential temperature on 18 August (see Fig. 2) are marked by a star symbol. End points in the BL [of and points of strongest updraft for the cirrus cloud observations](#) are found in different regions, similar as for the cirrus on
- 20 15 August, demonstrating that the air masses in the cirrus are a mixture of different origins as [well as found](#) in the ATAL itself.

As mentioned in Sect. 4.2, cirrus formation as well as likely the details of transport pathways that lead to cirrus formation are not represented in our CLaMS trajectory calculations.

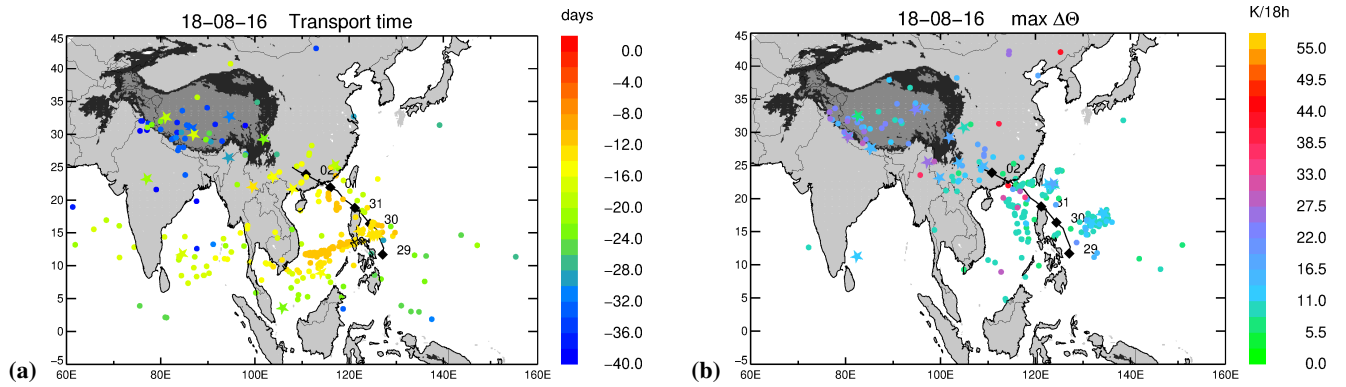


**Figure 10.** The same as Fig. 4 and Fig. 7, but for Case 3 the typhoon-influenced ATAL on 18 August 2016. The trajectories are calculated in a potential temperature range between 150 hPa to 75 hPa (362 K to 422 K).



**Figure 11.** The same as Fig. 5 and Fig. 8, but for the balloon flight on 18 August 2016 heavily influenced by a typhoon in the western Pacific (Case 3).





**Figure 12.** The same as [As](#) Fig. 6 and Fig. 9, but for the typhoon-influenced case on 18 August 2016 (Case 3). Locations of end points and locations of the strongest updraft of trajectories initialised within the cirrus cloud which was found within the ATAL are indicated by a star symbol. The 40-day backward trajectories are impacted by typhoon Nida. Its storm track is shown (black line) and the location at noon time is additionally marked (black diamonds); the numbers indicate the day in July and August 2019.

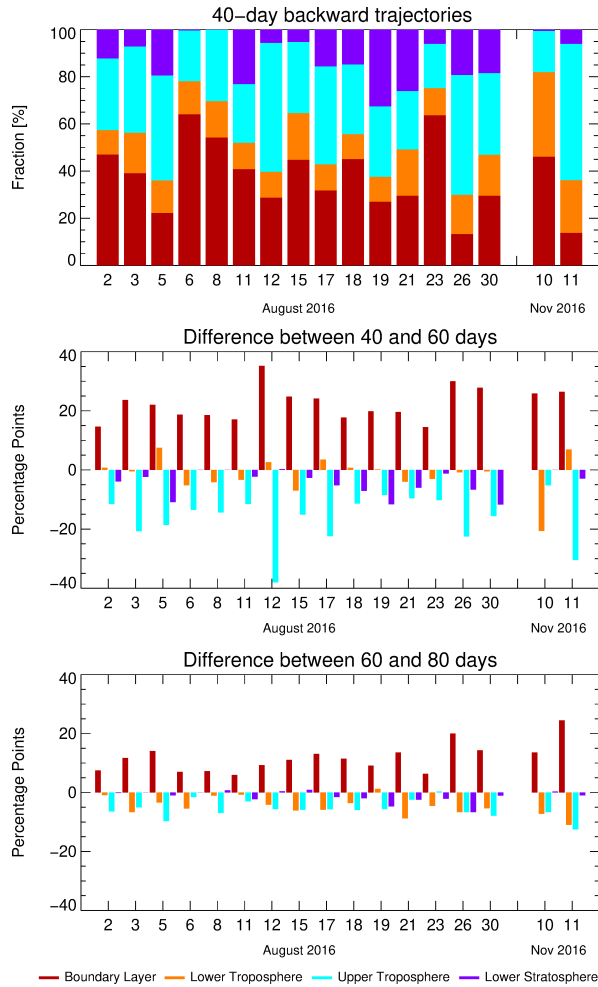
#### 4.4 Results for all flights

In Sects. 4.1, 4.2, and 4.3, the fractions of the different atmospheric height-altitude layers (BL, LT, UT, LS) contributing to the composition of air masses within the ATAL were discussed in detail for three specific days, namely the 6, 15 and 18 August 2016. In Fig. 13 (top), the fractions of the different atmospheric height-altitude layers for all flights in August and November 2016 are shown as a bar chart (see Tab. 1 and Tab. 2) for 40-day backward trajectories. Due to the strong variability of the vertical extent of the ATAL and the variability of cirrus clouds marking the bottom of the ATAL for certain days, the number of back-trajectories calculated for a particular day varies strongly from a ~~number of~~ 119 to up to 704. For better comparison the fractions of the different atmospheric height-altitude layers are normalised by the total number of trajectories for each day (Fig. 13, top). The fractions from the BL are between 14% and 64% and from the LS between 0% and 33%. Thus there is a strong variability of the fractions of the different atmospheric height-altitude layers contributing to the ATAL as well as for the no ATAL cases in August and post-monsoon cases in November within the ~~corresponding~~ pressure levels between 140 and 92 hPa. However, from the analysis of different atmospheric height-altitude layers no clear relation between these height-altitude layers and the occurrence of ATAL was found, that may explain the difference between the ATAL and no ATAL cases as well as between ATAL and post-monsoon soundings in November.

During the monsoon season air masses from the BL accumulate within the Asian monsoon anticyclone, thus in the altitude range of the ATAL. Therefore, back-trajectory calculations with a length of 60 and 80 days were also performed to consider the sensitivity of our results regarding on the trajectory length. In general the fractions from the BL contributing to the ATAL increase with trajectory time. The fractions from the BL range between 14% to 64% for 40 days (Fig. 13, top), from 40% to 83% for 60 days and from 56% to 90% for 80 days. There is an increase of the fractions from the BL between 14 and 36 percentage points between 40 and 60 days (Fig. 13, middle) and between 6 and 25 percentage points between 60 and 80 days (Fig. 13, bottom). Simultaneously the fractions from the UT decrease with time. They range between 17% to 58% for 40 days, from 8% to 28% for 60 days and from 6% to 21% for 80 days (see Fig. 13). A decrease is found of the fractions from the UT between 5 and 38 percentage points between 40 and 60 days (Fig. 13, middle) and between 0 and 12 percentage points between 60 and 80 days (Fig. 13, bottom). The fractions from the LT do not change significantly with increasing trajectory length. They vary between  $-7$  and  $8$  percentage points between 40 and 60 days (except for 10 November 2016, here the difference is  $-21$  percentage points) and between  $-11$  and  $1$  percentage points between 60 and 80 days. The fractions from the LS decrease by up to  $-11$  percentage points between 40 and 60 day and up to  $-7$  points between 60 and 80 days.

To deduce a possible relation between the ATAL intensity and the air mass origin within the boundary layer, Fig. 14a shows a bar chart with the contributions of Tibet, Foothills, Land, Ocean and Residual where the individual measurements are sorted by increasing mean backscatter ratio ( $\overline{\text{BSR}}_{455-1} \times 100$ ) (see Tabs. 1 and 3). The contributions are normalised by the total number of trajectories within the boundary layer for each day for better comparison. The balloon flight on 12 August is excluded because a cirrus cloud was detected between 140 and 92 hPa and therefore no aerosol backscatter ratio could be measured in this pressure range. Because of the low statistics we exclude all flights (11, 21, 23 and 26 August) in Fig. 14a where the





**Figure 13.** The fractions of the different atmospheric ~~height~~altitude layers (BL, LT, UT, LS) for all flights in August and November 2016 normalised by the total number of trajectories of each day calculated from 40-day backward trajectories (top) (see Tab. 2), the difference between 40 and 60 days (middle) backward trajectory calculations as well as between 60 and 80 days (bottom).

trajectory number is lower than 50% of the maximum number of trajectories (# 704) calculated on 18 August 2016 (however these cases are included in Fig. 15).

There is a lot of variability of the air mass origin within the boundary layer contributing to ATAL cases characterised by a backscatter ratio  $\overline{\text{BSR}}_{455}$  larger than 1.058 (Fig. 14, top). Strong ATAL cases with  $\overline{\text{BSR}}_{455}$  values larger than 1.067 show

5 fractions of air mass origin in the boundary layer from continental outflow higher than 70% and in particular higher than 30% from Tibet (Fig. 14, top). For weak ATAL cases (18 and 30 August), the  $\overline{\text{BSR}}_{455}$  values are lower (1.059–1.065) and the ~~fractions~~fraction from continental outflow is below 70%. For these cases high fractions from maritime boundary layer

sources (Ocean > 12%–30%) are found which are caused by the impact of tropical cyclones on the composition of the ATAL as discussed in Sect. 4.3.

For both post-monsoon cases in November 2016 low  $\overline{\text{BSR}}_{455}$  values of 1.034 and 1.037 are measured, these measurements are used as background signal for aerosols in the UTLS (during August 2016 (Fig. 1 shows that no ATAL has been observed during winter). For these cases the air mass origin is very different, compared to the ATAL cases. In 40-day back-trajectory calculations no contributions from Tibet and Foothills are found for both flights, only a contribution from Land of 4% is found for the balloon flight on 11 November 2016. The origins-origin of air masses in the boundary layer are from the Ocean and from Residual mainly from south of 5°S. It is known that during boreal winter efficient transport into the stratosphere is found over the west Pacific and Maritime Continent, caused by strong convection and in-addition-, in addition, by the ascending branch of the Walker circulation located over the Maritime Continent (e.g. Bergman et al., 2012; Hosking et al., 2012).

Case 2, the no ATAL case from 15 August 2016, has a  $\overline{\text{BSR}}_{455}$  value of 1.023 lower than both November cases, however an air mass origin in the boundary layer is found of around 80% from continental sources (Tibet, Foothills and Land). This BL fraction is similar to fractions from the BL as for the ATAL cases. Case 2 is discussed in detail in Sect. 4.2 and it was shown that on 15 August a bi-modal structure of the anticyclone is found and Nainital is located between the eastern and western part of the anticyclone (Fig. 3). This bi-modal structure is found between 11 and 16 August (not shown here). Thus also the no ATAL case on 12 August and the balloon-flight on 11 August are impacted by this dynamical situation in the UTLS. Therefore, the dynamics in the UTLS seems to be the reason that on 15 August 2016 no ATAL was measured over Nainital, although a similar air mass origin was found on 15 August as for the ATAL cases. Unfortunately, during August 2016 we have only one no ATAL measurement (not-completely-overlaid-which is not completely maked by a cirrus cloud), therefore we can not deduce a more general result regarding no ATAL measurements during the peak monsoon season in India in our study. During August 2016, a second period with a bi-modal structure of the anticyclone is found between 27 and 28 August, but here Nainital is located more in the centre of the eastern mode (not shown here).

~~The change between the ATAL intensity and the air mass origin within the boundary layer (Tibet, Foothills, Land, Ocean and Residual) between 40 and 60 days (Fig. 14b) as well as between 60 and 80 days (Fig. 14c) is also analysed. In general, the fractions of the Residual is increasing with the trajectory length for all balloon flights, except for 11 November. Thus the longer the trajectories, the more older air masses are taken into account to~~ However, we can not exclude that also aerosol removal processes depending on temperature and  $S_{\text{ice}}$  such as in-situ cirrus formation contribute to the ~~ATAL coming from outside the latitude-longitude box from 60°E to 160°E and from 5°S to 45°N. For strong ATAL cases, in general an increase of the fractions from Ocean up to ~7 percentage points are found (Fig. 14b) indicating the impact of maritime convection between 40 and 80 days before the soundings. In contrast, for weak ATAL cases, the fraction of continental convection (Tibet, Foothills, and Land) is increasing, indicating the impact of continental convection between 40 and 80 days before the soundings~~existence of the no ATAL measurement on 15 August 2016 (details see Sect. 5).

Figs.Figures 6, 9 and 12 show that on 6, 15 and 18 August the strongest updraft locations differ from the endpoint locations of the trajectories. A cluster of strongest updraft locations is found at the southern edge of the Himalayan foothills. Tab.Table 4 shows the fractions of Tibet, Foothills, Land, Ocean and Residual of the strongest updraft locations within 18 hours for all

flights. Our definition of the foothills at the southern edge of the Himalaya by the orography results in this region being only a small belt ~~identified by the orography~~. Thus not all trajectories having their strongest updraft at the southern edge of the Himalayan foothills are captured by our definition of the foothills and therefore these trajectories are ~~still~~ also counted as fractions of Land ~~Fig. or Tibet. Figure~~ 14d shows the difference between the location of the end points and the location of strongest updraft for 40-day backward calculations. For ~~some soundings (3, 5, 6, 8 and 30 August)~~ all ATAL soundings the fractions from Foothills (except on 6 August) and Tibet for the location of strongest updraft are greater than for the location of the end points indicating strong convective activity at the southern edge of the Himalayan foothills during early August 2016. On 3, 6 and 8 August, the fractions from Tibet are more than 10 percentage points larger for the the location of strongest updraft than for the location of the end points.

10 ~~Fig. Figure~~ 15 shows the fractions of Tibet, Foothills, Land and Ocean versus  $(\overline{\text{BSR}}_{455}-1) \times 100$  (see Tab. 1) for the location of the end points of the trajectories (~~top~~) (see Tab. 3) ~~and for the location of the strongest updraft (bottom) (see Tab. 4)~~. The fractions are not normalised to the number of trajectories within the BL as was done in Fig. 14. Therefore, in Fig. 15 also the contribution from the other atmospheric layers (LT, UT and LS) to the ATAL are taken into account. Further, the ~~low statistic flights~~ (flights with a relatively low absolute number of trajectories ~~7~~ (11, 21, 23 and 26 August) are also considered. There is

15 a lot of variability between individual soundings. However, in general an increase of the fractions from Tibet, Foothills and Land is found for increasing ATAL intensity, while the contributions from the Ocean are decreasing for ~~both~~ the location of end points ~~and the locations of strongest updraft. The gradient of the linear fit for the fractions~~.

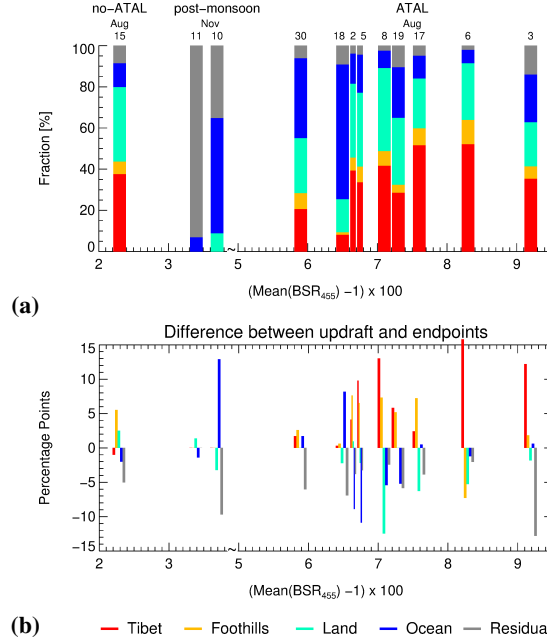
Further, the Pearson correlation coefficients between the different regions (Tibet, Foothills, Land and Ocean) and the backscatter intensity averaged over the ATAL altitude range  $((\overline{\text{BSR}}_{455}-1) \times 100)$  for the location of the end points of the

20 trajectories as well as for the location of the strongest updraft along the trajectories is calculated (see Tab. 5). Low positive correlations (i.e. values from 0.3 to 0.5) are found for the location of the end points of Tibet and Foothills ~~is somewhat steeper for the locations of strongest updraft compared to the location of the endpoints highlighting the role of the southern Himalayan Foothills and the Tibetan Plateau in the uplift of~~ as well as for the location of the strongest updraft of Tibet.

Moreover, the Pearson correlation coefficients are calculated excluding also the flights with a relatively low absolute number

25 of trajectories (11, 21, 23 and 26 August) and the no ATAL case on 15 August. The no ATAL case is excluded because the low backscatter intensity is not completely explained through the backward trajectory analysis presented here. Moderate positive correlations (i.e. values from 0.5 to 0.7) are found for Tibet, Foothills and Land with the largest correlation coefficient of 0.7 for the location of endpoints in Tibet. Low negative correlations (i.e. values from -0.3 to -0.5) are found for contributions from the Ocean. Overall, our findings show that the stronger the backscatter intensity averaged over the ATAL altitude range,

30 the higher the continental contributions from Tibet, Foothills and Land. The weaker the ATAL aerosols and their chemical precursors from the Earth's surface to ATAL altitudes is during August 2016, the higher are the maritime contributions.

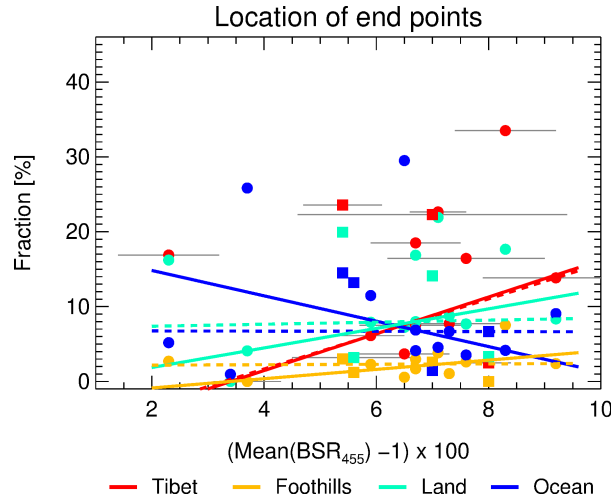


**Figure 14.** Contributions of Tibet, Foothills, Land, Ocean and Residual to the boundary layer (BL) sorted by increasing backscatter ratio  $(\overline{\text{BSR}}_{455} - 1) \times 100$  (see Tabs. 1 and 3) for 40-day backward trajectories (a). The contributions are normalised by the total number of trajectories within the boundary layer for each day. The days of the soundings in August and November are indicated above the top of the bars. For better representation, the range between 4.0 and 5.0 is removed from the x-axis. ~~Low-statistics~~ The flights (11, 21, 23 and 26 August) where the trajectory number is lower than 50% of the maximum number of trajectories (# 704) calculated on 18 August 2016 are not shown as well as the 12 August, when the UTLS is filled by a 5 km thick cirrus cloud. The difference between ~~40-and-60-day-and-between~~ ~~60-and-80-day-backward-trajectory-calculations-as-well-the-difference-between~~ the location of the end points and the location of strongest updraft (b) for 40-day backward calculations is also shown.

## 5 Discussion

COBALD measurements using the CI, the 940-to-455 nm ratio of the aerosol component of the BSR, can be used to discriminate aerosol and cirrus measurements (see Sect. 2.1). However, in case of a cirrus cloud, the dominant ice particle backscatter does not allow the COBALD sonde to detect, whether aerosol particles coexist with ice particles or not. Krämer et al. (2016) describe two types of cirrus: (1) in-situ origin cirrus observed at the altitudes where they are formed on soluble solution aerosol particles or on solid ice nucleating particles; (2) liquid origin, cirrus which are glaciated liquid clouds lifted from below to the cirrus temperature region. The formation mechanism of the cirrus cloud will have implications for the aerosol concentration in the cirrus layers within the ATAL.

For our back-trajectory calculations thin cirrus clouds within the ATAL are included to infer the origin of air masses contributing to the ATAL. If we assume that these cirrus clouds are of cirrus type (2), the aerosol concentration within the ATAL



**Figure 15.** Fractions of Tibet, Foothills, Land and Ocean versus  $(\overline{\text{BSR}}_{455} - 1) \times 100$  (see Tab. 1) for the location of the end points of the trajectories (top) (see Tab. 3) and for  $\overline{\text{BSR}}_{455}$  is the location of backscatter intensity averaged over the strongest updraft (bottom) ATAL altitude range listed in Tab. 1 for each flight (details see TabSect. 4.3.1). The standard deviation ( $\sigma$ ) of the backscatter ratio is indicated for each sounding (grey lines). Low-statistics The flights (11, 21, 23 and 26 August) where the trajectory number is lower than 50% of the maximum number of trajectories (#704) calculated on 18 August 2016 are also shown (indicated by squares instead of circles). For each region a linear fit (using the least absolute deviation method) was calculated using all measurements except the one on 12 August (Tab. 1), when the UTLS is filled by a 5 km km thick cirrus cloud (Tab dashed lines). Further, a linear fit excluding in addition the flights on 11, 21, 23 and 26 August as well as the no ATAL flight (15 August) is shown (lines). An increase of the fractions from Tibet, Foothills and Land is found for increasing ATAL intensity, while the contributions from the Ocean decrease for both the locations of end points and the locations of strongest updraft.

would not be affected by the formation process of the cirrus particles, however the uplift (convection) of air masses from the lower troposphere could transport enhanced concentrations of aerosol particles from close to the ground to ATAL altitudes (as proposed by Vernier et al., 2018). Vernier et al. (2018) found in some without much influence of aerosol in the middle troposphere (see Fig. 2). This concept is consistent with the results of Vernier et al. (2018), who found in OPC balloon-borne measurements of the ATAL from Hyderabad, India, in summer 2015 that aerosol concentrations in the boundary layer and in the ATAL are of the same magnitude, while much lower aerosol concentrations prevail in the middle troposphere. In their balloon measurements Vernier et al. (2018) further found that the presence of cirrus is associated with a reduction or a minimum in aerosol concentration possibly caused by aerosol removal processes through the formation of cirrus particles of type (1). Therefore, for both cirrus types, it is important to include the back-trajectory calculations of thin cirrus clouds found within the ATAL to identify the air mass origin of air masses contributing to the ATAL. Further, depending on the lifetime of the cirrus clouds within the ATAL, the occurrence of cirrus particles (e.g. through sedimentation or uptake on the ice surface) could have implication-implications on the ATAL even when the cirrus particles are no longer present.

Pearson Correlation Coefficients				
<u>location of</u>	<u>Tibet</u>	<u>Foothills</u>	<u>Land</u>	<u>Ocean</u>
<u>end points</u>	<u>0.33</u>	<u>0.35</u>	<u>0.14</u>	<u>-0.19</u>
<u>strongest updraft</u>	<u>0.41</u>	<u>0.09</u>	<u>-0.00</u>	<u>-0.19</u>
<u>end points*</u>	<u>0.70</u>	<u>0.63</u>	<u>0.56</u>	<u>-0.27</u>
<u>strongest updraft*</u>	<u>0.69</u>	<u>0.52</u>	<u>0.58</u>	<u>-0.32</u>

**Table 5.** Pearson correlation coefficients between the different regions (Tibet, Foothills, Land and Ocean) and the backscatter intensity averaged over the ATAL altitude range  $((BSR_{455}-1) \times 100)$  for the location of the end points of the trajectories (see Fig. 15) and for the location of the strongest updraft. For each region the Pearson correlation coefficient was calculated using all measurements except the one on 12 August, when the UTLS is filled by a 5 km thick cirrus cloud. Further, the Pearson correlation coefficients are also calculated excluding in addition low statistics flights (11, 21, 23 and 26 August) where the trajectory number is lower than 50% of the maximum number of trajectories (#704) calculated on 18 August 2016 (see Fig. 14) as well as the no ATAL case on 15 August (marked by \*).

We would like to emphasise that the calculation of  $\overline{BSR}_{455}$  (see Tab. 1) is based on binned data (with an altitude resolution of  $\sim 25$  m in the UTLS). ~~Small-Thin~~ cirrus clouds found within the ATAL are excluded for the calculation of the mean value of  $\overline{BSR}_{455}$ , because here the  $BSR_{455}$  value for cirrus is much higher than for aerosol (Sect. 3.1). However, the calculation of the fraction of air masses uses the high-resolution measurement data (with a resolution of 1 sec) for back-trajectories including the altitude ranges where ~~small-thin~~ cirrus clouds within the ATAL were detected. In the altitude range of ~~small-thin~~ cirrus clouds, the aerosol concentration could be enhanced by convection or reduced by aerosol removal processes such as in-situ cirrus formation. In both cases it is important to include the air masses in the altitude range where thin cirrus clouds were detected to identify the origin of air masses contributing to the ATAL.

However, cirrus clouds detected directly below the ATAL are excluded in our trajectory analysis, even if aerosol and cirrus coexisted here the bottom of the ATAL is highly uncertain. Therefore, in our analysis there is an uncertainty in the vertical altitude range of the ATAL. To solve this uncertainty balloon-borne measurements in addition to COBALD would be required such as the measurements of the particle size distribution (e.g. using an optical particle spectrometer, POPS; Gao et al. (2016)).

## 6 Conclusions

We presented a series of balloon-borne measurements performed by the COBALD instrument conducted in Nainital, Northern India, during August and November 2016. The  $BSR_{455}$  measurements show a strong day-to-day variability of the altitude, the vertical extent and the backscatter intensity of the ATAL at UTLS altitudes over Nainital in August 2016. ~~This variability is not visible in the climatological~~ In contrast to high-resolution in situ measurements, space-borne observations often do not allow studying the day-to-day variability of ATAL profiles therefore mean values of the ATAL ~~derived by profiles were frequently used in previous studies based on~~ satellite observations. (e.g. Vernier et al., 2011, 2015).

Further, there are frequent observations of cirrus layers embedded in the ATAL; depending on the duration and formation mechanism of the cirrus layers, there will be an impact of cirrus on the properties of the aerosol particles constituting the ATAL. In general the BSR<sub>455</sub> values measured in August are higher than values measured during post-monsoon in November, which represent the aerosol background during winter. However, there is one observation on 15 August showing no ATAL (here the BSR<sub>455</sub> is lower than in November).

Lagrangian back-trajectory calculations were performed using the CLaMS model driven by ERA-Interim reanalysis to identify the air mass origin in the model boundary layer and the transport pathways of air parcels contributing to the ATAL over Nainital in August 2016. ~~There~~ In this work, we show that there is a variety of factors impacting the variability of the ATAL: continental convection, tropical cyclones (maritime convection), dynamics of the anticyclone and stratospheric intrusions. All these factors contribute to the observed day-to-day variability of the ATAL found over Nainital in August 2016. Clearly the details of the vertical transport will differ, when a higher resolution, more recent reanalysis data set would be used, however the general transport patterns will likely remain unaltered. For example, the convective upward transport by tropical cyclones is found consistently for ERA-Interim and ERA5 in diabatic calculations (Li et al., 2020). Further, the ATAL is also impacted by other processes related to the formation and growth of aerosol as well as by aerosol removal processes such as in-situ cirrus formation.

We found that air masses contributing to the ATAL are a mixture of air masses from different origins and not exclusively from the BL. The trajectories originating from the BL are separated according to geopotential height and further, for low geopotential heights, between continental and maritime sources. Strong ATAL cases have high contributions from the Tibetan Plateau (Tibet), the remaining continental (Land) area and the foothills of the Himalayas (Foothills). Weaker ATAL cases have higher contributions from the maritime area (Ocean) caused by the impact of tropical cyclones. Here, mixing with unpolluted air masses from the maritime boundary layer yield a dilution of the ATAL. The no ATAL case of the 15 August 2016 has similar contributions from the Tibetan Plateau (Tib), the remaining continental (Land) area and the foothills of the Himalayas (Foothills) as the strong ATAL cases, however on this day there is a bi-modal structure of the anticyclone and Nainital is located between the western and eastern mode of Asian monsoon anticyclone. We hypothesise that the main driver for no ATAL cases ~~are~~ during the monsoon season is the dynamics of the Asian monsoon anticyclone. However, we have only one no ATAL measurement in August 2016 (partly influenced by cirrus), therefore ~~more~~ we can not exclude that also aerosol removal processes such as in-situ cirrus formation contribute to the existence of no ATAL measurements. More balloon-borne measurements ~~are necessary~~ would be required to validate this hypothesis.

Moreover, we calculated the locations of strongest updraft along the backward trajectories within 18 hours and found a cluster of such locations at the southern edge of the Himalayan foothills over northern India, Nepal and Bhutan. Further for balloon-measurements impacted by tropical cyclones locations of strongest updraft are also found over the western Pacific ocean. Within the ATAL, a mixture of air parcels with different transport times is found. The transport times from the Earth's surface to ATAL altitudes vary strongly. The shortest transport times found for 6, 15, and 18 August are between 10 and 15 days. These trajectories are originating either on the Tibetan Plateau or at the location of tropical cyclones in the western

Pacific. Shorter transport times below 10 days are only found for the 12 August (when the UTLS is filled by a ~~5km~~-km thick cirrus cloud) and 17 August for air masses originating on the Tibetan Plateau.

Finally, CLaMS backward trajectory calculations ~~identify~~-allow the transport pathways from the Earth's surface to ATAL altitudes to be identified. Very fast uplift in a convective range transports air masses up to the top of the convective outflow level (5 (~360 K) within a few days. Subsequently, the air parcels are slowly uplifted by diabatic heating within a large-scale upward spiral driven by the anticyclonic flow in the UTLS over the Asian monsoon region from about 360 K up to ATAL altitudes. Over Nainital in summer 2016, a maximum ATAL altitude of 422 K (75 hPa) was measured. This slow uplift caused by diabatic heating in a large-scale upward spiral is consistent with concepts referred to as 'an upward spiralling range' (Vogel et al., 2016) or as 'a confined lower stratosphere' (Brunamonti et al., 2018).

10 Our study contributes to deducing the source regions of emissions of precursors of ATAL particles at the Earth's surface and their transport pathways to the UTLS which is important to develop recommendations for regulations of anthropogenic surface emissions of ATAL precursors. In a recent study, Fadnavis et al. (2019b) argue that further increasing industrial emissions in Asia will lead to a wider and thicker ATAL having the potential to amplify the severity of droughts in India. Severe droughts would have fatal consequences on agriculture on the Indian subcontinent and therefore would result in strong socio-  
15 economic impacts in one of the most densely populated parts of the world. On the other hand, the ATAL impacts the radiative balance of the Earth's atmosphere and could have positive side effects in terms of reducing Earth's surface temperatures (Fadnavis et al., 2019b), therefore more future research about the ATAL and its impacts is required.

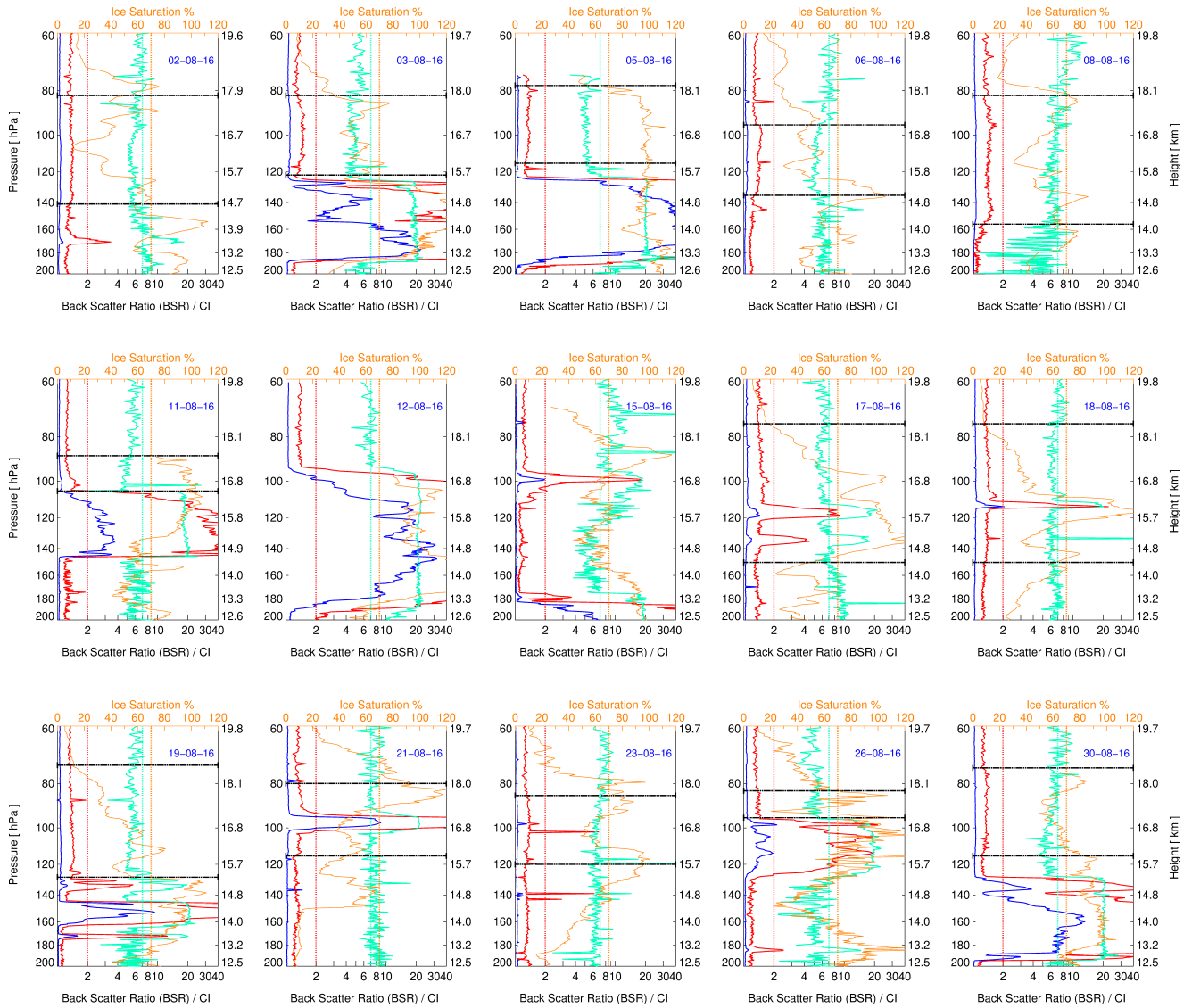
*Code and data availability.* The CLaMS code is available at the GitLab server: <https://jugit.fz-juelich.de/clams/CLaMS> (last access: 20 June 2019) as well as at the on-line repository Zenodo via <https://doi.org/10.5281/zenodo.2632683>. The typhoon tracks can be obtained at  
20 <https://www.jma.go.jp/jma/eng/jma-center/rsmc-hp-pub-eg/besttrack.html>. The data of the balloon sounding in Nainital 2016 used in this paper are available upon request from Sreeharsha Hanumanthu (s.hanumanthu@fz-juelich.de).

## 7 ~~Colour-Index CI~~

### Appendix A: Colour Index CI

During the balloon ascents from Nainital in August and November 2016 measurements were performed of backscatter at  
25 wavelengths of 455 nm (blue visible) and 940 nm (Wienhold, 2008; Brabec et al., 2012) and of water vapour mixing ratio by a cryogenic frost-point hygrometer (Vömel et al., 2007, 2016) (see section 2.1 in the main paper). These measurements allow the definition of a colour index (CI) (Cirisan et al., 2014) and the calculation of the ice saturation ( $S_{ice}$ ) during the balloon ascent. These quantities are considered in the analysis presented here to discriminate aerosol and cirrus clouds (ice particles) in the backscatter measurements and are shown in Fig. A1.



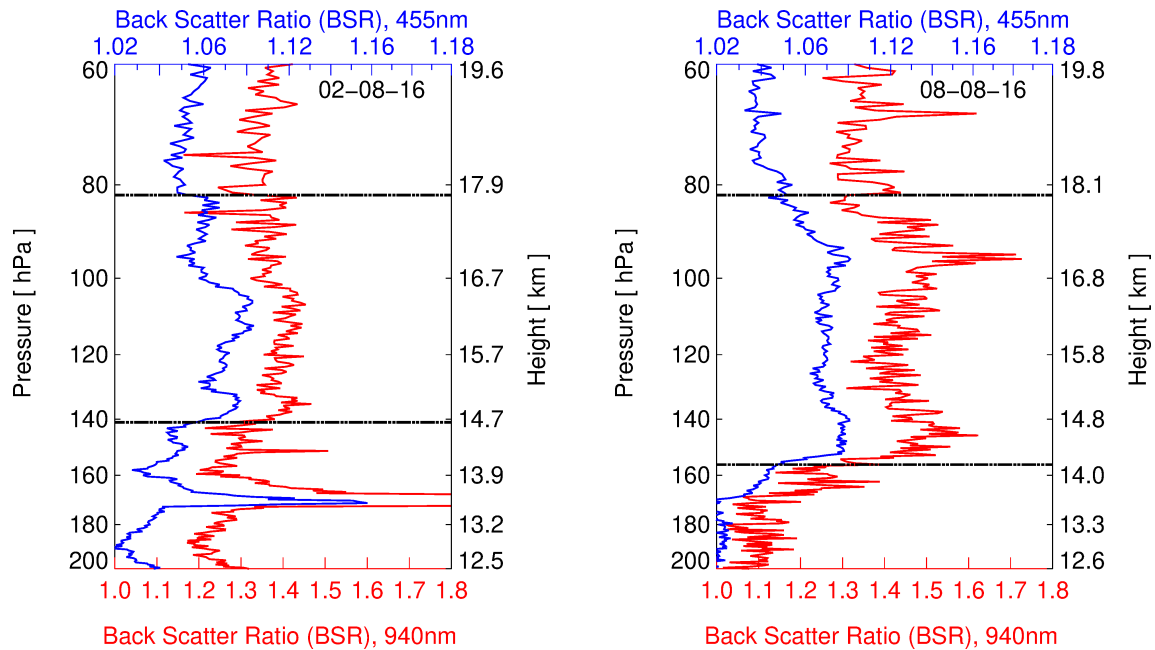


**Figure A1.** COBALD BSR<sub>455</sub> (blue) and BSR<sub>940</sub> (red) soundings in the UTLS region versus pressure 60–200 hPa for each flight in August 2016. In addition, the colour index ( $CI = (BSR_{940} - 1)/(BSR_{455} - 1)$ ; green) and the ice saturation ( $S_{ice}$ , orange) is shown. Corresponding height calculated using both measured pressure and temperature is also given for orientation (see right-hand y-axis). In our analysis, layers with  $CI > 7.0$  ( $CI = 7$ ; dotted green line),  $BSR_{940} \geq 2$  ( $BSR_{940} = 2$ ; dotted red line) and  $S_{ice} > 70\%$  ( $S_{ice} = 0.7$ ; dotted orange line) were rejected as cirrus clouds. The top and bottom boundaries of the ATAL (see Tab. 1) are marked by horizontal dashed black lines, except for 12 and 15 August, as no ATAL was detected on these days (as in Fig. 2).

## Appendix B: The Using the shortwave and longwave channel to detect the ATAL

The aerosol measurements analysed here are based on balloon-borne backscatter measurements employing the COBALD detector, which operates at optical wavelengths of 455 nm (blue visible) and 940 nm (infrared) (Brabec et al., 2012, see main paper) (Brabec et al., 2012). Commonly, the colour index (CI) is defined as the 940-to-455 nm ratio of the aerosol component of the BSR (Cirisan et al., 2014; Brunamonti et al., 2018, see also main paper) and provides some estimate of particle size. The BSR for both wavelength-wavelengths will give an indication of the presence of the ATAL. The same is true for cirrus particles, i.e., they can be detected by both the blue and the red channel, individually. But only by using the CI a clear discrimination of ice (CI > 7) and aerosol (CI < 7) is possible (see main paper), which is the advantage of measuring at two wavelengths.

Here and in previous work (Vernier et al., 2015, 2018; Brunamonti et al., 2018) the ATAL analysis is based on the 455 nm BSR measurements. Indeed, the 455 nm BSR measurement is the preferred channel for the detection of the ATAL. The main reason is that the 455 nm COBALD BSR measurement has a better precision and has a higher signal-to-noise ratio (although BSR is lower at 455 nm than 940 nm, the raw signal is higher at 455 nm). Further, the accuracy is better for 455 nm, as the scale parameter in the data processing is taken directly from the sonde profile. The 940 nm channel builds on that and the BSR values are restricted by the assumption that the CI has to remain in a certain range.



**Figure B1.** COBALD  $BSR_{455}$  (blue) and  $BSR_{940}$  (red) measurements in the UTLS region versus pressure 60-200 hPa for two flights in August 2016 (2 August, left panel, and 8 August, right panel). The right hand axis in both panels shows log-pressure height. Shown are the measured data in pressure bins without further processing (see main paper). The top and bottom boundaries of the ATAL (see Table 1 in the main paper) are marked by horizontal dashed-dot black lines. (Note the different scaling for  $BSR_{455}$  and  $BSR_{940}$ .)

In Fig. B1 we show the measured  $BSR_{455}$  (blue line) and  $BSR_{940}$  (red line) profiles from the COBALD measurements in Nainital in 2016 for two days in August (with no interference from cirrus layers or spike artefacts). The discussed features are clearly visible; although the ATAL can be detected using both the  $BSR_{455}$  and  $BSR_{940}$  measurements, the 455 nm channel is less noisy, so it constitutes the preferred channel for the detection of the ATAL in COBALD balloon measurements.

## 5 Appendix C: Removal of ‘moon spikes’

The processing of COBALD includes the rejection of ‘moon spikes’ that may arise ~~due to~~ during the night flights because of the oscillatory motion of the payload 60 m below the balloon, when the detector happens to be pointing towards the moon. Moon spikes affect only a tiny fraction of the COBALD data and care is taken not to confuse them with thin cirrus clouds.

To identify those anomalous ‘spikes’, we used a simple criterion; we consider the signal to be an anomalous spike when

10  $BSR_{455} \text{ nm} > 1.12$  and  $CI < 7$ . These spikes are then removed from the data set, where the condition  $CI < 7$  ensures that cirrus clouds will be retained.

Finally, some specific cases with spikes in the  $BSR_{455}$  measurements of a very small vertical extent still remained in the binned data, where  $BSR_{455} < 1.12$  and  $7 < CI < 10$ . These specific cases occurred for 3, 15, 21, 23, and 30 August; the corresponding data points were also removed from the data set used here.

15 *Author contributions.* S.B., T.J. and T.P. coordinated all measurements. The study was conceived by B.V., S.H. and R.M.. The measurements in Nainital were conducted by S.H., S.B., T.J., P.Ö., M.N., B.B.S., K.K., S.S. and T.P.. B.B.S., M.N., S.S. and S.F. provided logistic support for the measurements in Nainital. The trajectory calculations and the associated analysis were conducted by S.H. and B.V., with contributions by D.L.. The results of the study were discussed by all coauthors, with particular contributions by S.B., B.L., T.P., S.H., B.V., and R.M.. The paper was written by B.V., S.H., and R.M., with contributions from all coauthors.

20 *Competing interests.* The authors declare that no competing interests are present.

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