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.

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: $17km \rightarrow 18km$

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 cm⁻³) are dominated by particles in the $r > 0.094 \,\mu$ m channel, whereas particle number concentrations for the $r > 0.15 \,\mu$ m channel, and the $r > 0.30 \,\mu$ m channel are lower by a factor of 30 and 300, respectively (Vernier et al., 2018, Fig. 9).

6. P4L3-4: 0.6W/m2 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 ≈ 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 W/m^2 – compensating about one third of the comparable radiative forcing associated with the global increase in CO₂ (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 ~ 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/m². 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: Summertime nitrate aerosol in the upper troposphere and lower stratosphere over the Tibetan Plateau and the South Asian summer mon- soon 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, BSR940>2 AND Sice>70% \rightarrow OR

We have replaced the following text in the ACPD version:

'We reject layers with CI > 7.0, $BSR_{940} \ge 2$ 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 BSR₉₄₀ \geq 2 as well as an enhanced ice saturation (Sice > 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).

P6L30: Aerosol layers without cirrus can exist under the condition Sice > 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 BSR₄₅₅ 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, theres 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 (BSR₄₅₅) 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 Sice 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 that 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.

References

- Brunamonti, S., Jorge, T., Oelsner, P., Hanumanthu, S., Singh, B. B., Kumar, K. R., Sonbawne, S., Meier, S., Singh, D., Wienhold, F. G., Luo, B. P., Boettcher, M., Poltera, Y., Jauhiainen, H., Kayastha, R., Karmacharya, J., Dirksen, R., Naja, M., Rex, M., Fadnavis, S., and Peter, T.: Balloon-borne measurements of temperature, water vapor, ozone and aerosol backscatter on the southern slopes of the Himalayas during StratoClim 2016–2017, Atmos. Chem. Phys., 18, 15937–15957, https://doi.org/10.5194/acp-18-15937-2018, URL https://www.atmos-chem-phys.net/18/15937/2018/, 2018.
- Deshler, T., Hervig, M. E., Hofmann, D. J., Rosen, J. M., and Liley, J. B.: Thirty years of in situ stratospheric aerosol size distribution measurements from Laramie, Wyoming (41 degrees N), using balloon-borne instruments, J. Geophys. Res., 108, 4167, https://doi.org/10.1029/2002JD002514, 2003.
- Fadnavis, S., Semeniuk, K., Pozzoli, L., Schultz, M. G., Ghude, S. D., Das, S., and Kakatkar, R.: Transport of aerosols into the UTLS and their impact on the Asian monsoon region as seen in a global model simulation, Atmos. Chem. Phys., 13, 8771–8786, https://doi.org/10.5194/acp-13-8771-2013, 2013.
- Fadnavis, S., Sabin, T. P., Roy, C., Rowlinson, M., Rap, A., Vernier, J.-P., and Sioris, C. E.: Elevated aerosol layer over South Asia worsens the Indian drought, Sci. Rep., 9, 10268, https://doi.org/10.1038/s41598-019-46704-9, 2019.

- Fairlie, D., Vernier, J.-P., Natarajan, M., and Bedka, K. M.: Dispersion of the Nabro volcanic plume and its relation to the Asian summer monsoon, Atmos. Chem. Phys., 14, 7045–7057, https://doi.org/10.5194/acp-14-7045-2014, 2014.
- Fairlie, T. D., Liu, H., Vernier, J.-P., Campuzano-Jost, P., Jimenez, J. L., Jo, D. S., Zhang, B., Natarajan, M., Avery, M. A., and Huey, G.: Estimates of Regional Source Contributions to the Asian Tropopause Aerosol Layer Using a Chemical Transport Model, Journal of Geophysical Research: Atmospheres, 125, e2019JD031506, https://doi.org/10.1029/2019JD031506, URL https://agupubs.onlinelibrary.wiley.com/doi/abs/10.1029/ 2019JD031506, e2019JD031506 2019JD031506, 2020.
- Gao, R. S., Telg, H., McLaughlin, R. J., Ciciora, S. J., Watts, L. A., Richardson, M. S., Schwarz, J. P., Perring, A. E., Thornberry, T. D., Rollins, A. W., Markovic, M. Z., Bates, T. S., Johnson, J. E., and Fahey, D. W.: A light-weight, high-sensitivity particle spectrometer for PM2.5 aerosol measurements, Aerosol Science and Technology, 50, 88– 99, https://doi.org/10.1080/02786826.2015.1131809, URL https://doi. org/10.1080/02786826.2015.1131809, 2016.
- Gu, Y., Liao, H., and Bian, J.: Summertime 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, https://doi.org/ 10.5194/acp-16-6641-2016, URL https://www.atmos-chem-phys.net/ 16/6641/2016/, 2016.
- Hoskins, B. J. and Rodwell, M. J.: A model of the Asian summer monsoon, I, The global scale, J. Atmos. Sci., 52, 1329–1340, 1995.
- Lau, W. K. M., Yuan, C., and Li, Z.: Origin, Maintenance and Variability of the Asian Tropopause Aerosol Layer (ATAL): The Roles of Monsoon Dynamics, Sci. Rep., 8, 3960, 2018.
- Li, D., Vogel, B., Müller, R., Bian, J., Günther, G., Li, Q., Zhang, J., Bai, Z., Vömel, H., and Riese, M.: High tropospheric ozone in Lhasa within the Asian summer monsoon anticyclone in 2013: influence of convective transport and stratospheric intrusions, Atmos. Chem. Phys., 18, 17979–17994, https://doi.org/10.5194/acp-18-17979-2018, URL https:// www.atmos-chem-phys.net/18/17979/2018/, 2018.

- Pan, L. L., Paulik, L. C., Honomichl, S. B., Munchak, L. A., Bian, J., Selkirk, H. B., and Vömel, H.: Identification of the tropical tropopause transition layer using the ozone-water vapor relationship, Journal of Geophysical Research: Atmospheres, 119, 3586–3599, https://doi.org/10.1002/ 2013JD020558, URL https://agupubs.onlinelibrary.wiley.com/doi/ abs/10.1002/2013JD020558, 2014.
- Randel, W. J. and Park, M.: Deep convective influence on the Asian summer monsoon anticyclone and associated tracer variability observed with Atmospheric Infrared Sounder (AIRS), J. Geophys. Res., 111, D12314, https://doi.org/ 10.1029/2005JD006490, 2006.
- Ridley, D. A., Solomon, S., Barnes, J. E., Burlakov, V. D., Deshler, T., Dolgii, S. I., Herber, A. B., Nagai, T., Neely III, R. R., Nevzorov, A. V., Ritter, C., Sakai, T., Santer, B. D., Sato, M., Schmidt, A., Uchino, O., and Vernier, J. P.: Total volcanic stratospheric aerosol optical depths and implications for global climate change, Geophys. Res. Lett., 41, 7763–7769, https://doi.org/ 10.1002/2014GL061541, URL https://agupubs.onlinelibrary.wiley. com/doi/abs/10.1002/2014GL061541, 2014.
- Thomason, L. W. and Vernier, J.-P.: Improved SAGE II cloud/aerosol categorization and observations of the Asian tropopause aerosol layer: 1989–2005, Atmos. Chem. Phys., 13, 4605–4616, https://doi.org/10.5194/acp-13-4605-2013, URL https://www.atmos-chem-phys.net/13/4605/2013/, 2013.
- Turner, A. G. and Annamalai, H.: Climate change and the South Asian summer monsoon, Nature Climate Change, 2, 587–595, https://doi.org/10.1038/ nclimate1495, URL https://doi.org/10.1038/nclimate1495, 2012.
- Vernier, J. P., Fairlie, T. D., Natarajan, M., Wienhold, F. G., Bian, J., Martinsson, B. G., Crumeyrolle, S., Thomason, L. W., and Bedka, K. M.: Increase in upper tropospheric and lower stratospheric aerosol levels and its potential connection with Asian pollution, J. Geophys. Res., 120, 1608–1619, https://doi.org/ 10.1002/2014JD022372, 2015.
- Vernier, J.-P., Fairlie, T. D., Deshler, T., Ratnam, M. V., Gadhavi, H., Kumar, B. S., Natarajan, M., Pandit, A. K., Raj, S. T. A., Kumar, A. H., Jayaraman, A., Singh, A. K., Rastogi, N., Sinha, P. R., Kumar, S., Tiwari, S., Wegner, T., Baker, N., Vignelles, D., Stenchikov, G., Shevchenko, I., Smith, J., Bedka, K., Kesarkar, A., Singh, V., Bhate, J., Ravikiran, V., Rao, M. D., Ravindrababu, S., Patel,

A., Vernier, H., Wienhold, F. G., Liu, H., Knepp, T. N., Thomason, L., Crawford, J., Ziemba, L., Moore, J., Crumeyrolle, S., Williamson, M., Berthet, G., Jégou, F., and Renard, J.-B.: BATAL: The Balloon Measurement Campaigns of the Asian Tropopause Aerosol Layer, Bull. Am. Meteorol. Soc., 99, 955–973, https://doi.org/10.1175/BAMS-D-17-0014.1, 2018.

- Vogel, B., Müller, R., Günther, G., Spang, R., Hanumanthu, S., Li, D., Riese, M., and Stiller, G. P.: Lagrangian simulations of the transport of young air masses to the top of the Asian monsoon anticyclone and into the tropical pipe, Atmos. Chem. Phys., 19, 6007–6034, https://doi.org/10.5194/acp-19-6007-2019, URL https://www.atmos-chem-phys.net/19/6007/2019/, 2019.
- Yu, P., Rosenlof, K. H., Liu, S., Telg, H., Thornberry, T. D., Rollins, A. W., Portmann, R. W., Bai, Z., Ray, E. A., Duan, Y., Pan, L. L., Toon, O. B., Bian, J., and Gao, R.-S.: Efficient transport of tropospheric aerosol into the stratosphere via the Asian summer monsoon anticyclone, Proc. Natl. Acad. Sci., 114, 6972–6977, https://doi.org/10.1073/pnas.1701170114, URL https: //www.pnas.org/content/114/27/6972, 2017.
- Yuan, C., Lau, W. K. M., Li, Z., and Cribb, M.: Relationship between Asian monsoon strength and transport of surface aerosols to the Asian Tropopause Aerosol Layer (ATAL): interannual variability and decadal changes, Atmos. Chem. Phys., 19, 1901–1913, https://doi.org/10.5194/acp-19-1901-2019, URL https://www.atmos-chem-phys.net/19/1901/2019/, 2019.