Dear Referee 1,
We thank you a lot for your valuable comments and suggestions. We addressed them as explained below.
The reviewer's comments are repeated in **bold letters**, our replies are given in standard font, and text modified or added to the manuscript is given in blue.

In this study, the authors investigate the importance of aerosol dynamics and aerosol-radiation interactions in the early dispersion of the volcanic plume injected by the Raikoke eruption in June 2019. They argue that physical processes influencing the transport of volcanic plumes in the UTLS region have been poorly addressed compared to work related to source parameters/initial conditions. Using a set of satellite observations including HIMAWARI-8, CALIOP and OMPS-LP, they attempt to validate their simulations of the ICON-CART global modelling system. This is a very interesting and unique study that attempt to shed light on how a complex aerosol-dynamic-radiation coupling system can be used to understand early evolution of volcanic plumes and thus is suitable for publication in the Atmospheric Chemistry and Physics Journal. However, I believe that additional work would need to be done to validate the model results. With only one CALIPSO browse image and one OMPS-LP volcanic plume top point, the vertically resolved information that offer a unique opportunity to validate model results are not fully explored. Before this manuscript can be published, I would recommend the authors to provide additional observational evidences to support their conclusions.

Thank you very much for the insightful review. Your comments and questions helped us a lot to improve the manuscript.
We agree that additional observational data, especially in form of OMPS-LP volcanic cloud top height, would be very beneficial for the validation of the model results. Unfortunately, we were not able to retrieve any meaningful volcanic cloud height from OMPS-LP measurements for other dates. The reason for this is discussed in more detail together with the answer to comment 7.
The CALIOP measurements show a signal that can be associated with the volcanic cloud on other dates as well. We included these in our answers to the respective comments.

1. P1L3: I agree with this statement but essential information about mass injection rates and plume injection heights are still critical parameters to simulate volcanic plume dispersion.

Yes, we totally agree with you that the correct representation of source parameters is very critical for a reliable forecast of volcanic aerosols. Especially, estimates about the mass eruption rate and plume height are crucial for short term forecasts right after volcanic eruptions. This is why they are still substance of ongoing research. With this work, we don't intend to diminish the importance of source parameters, but shed light on less studied sink processes.

2. P1L10: I would replace "show" by "suggest" since I'm not certain that the results presented in this paper really fully support the conclusions.

During the review process we had the opportunity to provide further evidence for our statement. This is why we would like to leave it as is.

3. P2L36: I would argue that the rise of the plume is better documented by the two initial papers from Khaykin et al., 2017 and Peterson et al., 2017.

We additionally cite the suggested two papers:

Khaykin, S. M., Godin-Beekmann, S., Keckhut, P., Hauchecorne, A., Jumelet, J., Vernier, J.-P., Bourassa, A., Degenstein, D. A., Rieger, L. A., Bingen, C., Vanhellemont, F., Robert, C., DeLand, M., and Bhartia, P. K.: Variability and evolution of the midlatitude stratospheric aerosol budget from 22 years of ground-based lidar and satellite observations, Atmos. Chem. Phys., 17, 1829–1845, https://doi.org/10.5194/acp-17-1829-2017, 2017.

Peterson, P. K., Pöhler, D., Sihler, H., Zielcke, J., General, S., Frieß, U., Platt, U., Simpson, W. R., Nghiem, S. V., Shepson, P. B., Stirm, B. H., Dhaniyala, S., Wagner, T., Caulton, D. R., Fuentes, J. D., and Pratt, K. A.: Observations of bromine monoxide transport in the Arctic sustained on aerosol particles, Atmos. Chem. Phys., 17, 7567–7579, https://doi.org/10.5194/acp-17-7567-2017, 2017.

We add to the manuscript in l. 31:
This can result in a lofting mechanism of aerosol which is different from the one caused by large scale atmospheric dynamics as described for example by Khaykin et al. (2017).

In l. 34:
Peterson et al. (2017) observed in the Arctic near-surface atmosphere that the transport of atmospheric pollutants is influenced by active halogen chemistry.

4. P3L83: Could you explain what's the implications of selecting qa_value larger than 0.5?

The qa_value is described in the ESA Tropomi User Manual as followed:
"The quality value or qa_value is a continuous quality descriptor, varying between 0 (no data) and 1 (full quality data). Recommend to ignore data with qa_value < 0.5 (static)" (Sentinel-5 precursor/TROPOMI Level 2 Product User Manual Sulphur Dioxide SO2, https://sentinel.esa.int/documents/247904/2474726/Sentinel-5P-Level-2-Product-User-Manual-Sulphur-Dioxide , accessed 23 July 2020)
In order to improve comprehensibility, we reformulate P3L83
Only data with the quality descriptor 'qa_value' larger than 0.5 and total vertical column density values less than 1000 mol m^{-2} were used.

to:
Only data with a quality value larger than 0.5 (as recommended in the TROPOMI product user manual) and total vertical column density with values less than 1000 mol m^{-2} were used.

5. P4L109: One sentence about the adjustment technique could be explained here.

We rephrase and add some extra information in l.109.
Water vapor and clouds cause interference with the SO_2 signal and introduce a positive bias. Therefore, a retrieval scheme was devised to minimize the interfering effects. In short, the bias is minimized by subtracting an offset SO_2 retrieval for a small region where no SO_2 is believed to exist.

6. P5L126: What could be the impact of ice on those estimates?

We thank the reviewer for raising this as it is a very good point and one that we should have addressed in the manuscript. Ice formation in volcanic clouds is a known problem and happens often, especially in water-rich and tropical eruptions where moist air entrainment happens see Prata et al. (2020). Ice has a very clear infrared spectral signature that can be used to diagnose its presence in volcanic clouds. For Raikoke this signature was absent or at best, weak. True-color images from the Himawari-8 satellite also show no obvious signs of ice - the clouds are dark brown and become paler with time, presumably because of dispersion. The absence of an ice signature can be explained by the high altitude of the emissions (>8 km and up to 15 km) which deposited them into a very dry part of the atmosphere, and the lack of a water-rich plume to begin with, as evidenced in the true-color and IR spectral signature data. The presence of ice reduces the ash mass estimates by an amount that depends on the proportion of the pixel covered by ice. Ice formation could have occurred in the early (first few hours of the eruption on 21 June) as the plume ascended through a moister part of the atmosphere. This could partly explain why ash estimates at the start of the eruption are low; but ash opacity is also a factor that reduces the ash mass retrieval.

Prata, A.T., Folch, A., Prata, A.J., Biondi, R., Brenot, H., Cimarelli, C., Corradini, S., Lapierre, J. and Costa, A., 2020. Anak Krakatau triggers volcanic freezer in the upper troposphere. Scientific reports, 10(1), pp.1-13.

In order to address this issue, we add the following to the manuscript at l.120:

The presence of ice reduces the ash mass estimates by an amount that depends on the proportion of the pixel covered by ice. However, during the Raikoke eruption, ice was not observed except possibly at the start of the eruption which could cause lower ash mass estimates.

7. P6L167: This is very unlikely that the Ambae eruption had a significant impact on stratospheric aerosols beyond the tropics and sub-tropics and thus it seems unrealistic to consider that Ambae could impact the retrieval of a fresh volcanic plume within the OMPS data set within the latitude band where the Raikoke was transported during the first few days.

As the study of Malinina et al. (2020, in review at ACP) on the Ambae eruption shows, the Ambae plume spreads up to 40N until the end of 2018, where its influence still remains non-negligible. Thus, influence of the Ambae eruption at 50N in June 2019 might be expected. We agree with the reviewer that these small residual signals do not affect the retrievals in the core of the fresh Raikoke plume. Unfortunately, the sampling of the OMPS-LP instrument is quite sparse and it does not hit the core of the fresh plume on other days than the one analyzed in the paper. During the analyzed period, in the transition regions of the plume the increase of the aerosol extinction measured by OMPS-LP was not that pronounced and thus can be interfered by residual signals from previous events. We change the manuscript to make this clearer.

Malinina, E., Rozanov, A., Niemeier, U., Peglow, S., Arosio, C., Wrana, F., Timmreck, C., von Savigny, C., and Burrows, J. P.: Changes in stratospheric aerosol extinction coefficient after the 2018 Ambae eruption as seen by OMPS-LP and ECHAM5-HAM, Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2020-749, in review, 2020

We additionally cite Malinina et al. (2020) in l. 161:
Detailed information on the retrieval algorithm can be found in Malinina (2019) and Malinina et al. (2020).

We rephrase l. 165ff.:
In the following days, when the plume started to spread over the North Pacific, the core of the fresh plume is not hit by the OMPS-LP instrument sampling anymore. Slightly perturbed aerosol extinction observed in transition regions has a similar magnitude as that from interfering events, e.g., the aerosol transport from the Ambae eruption that occurred 11 months earlier, and thus cannot be attributed exclusively to the Raikoke eruption. For this reason, we excluded the OMPS-LP measurements in transition regions from the consideration.

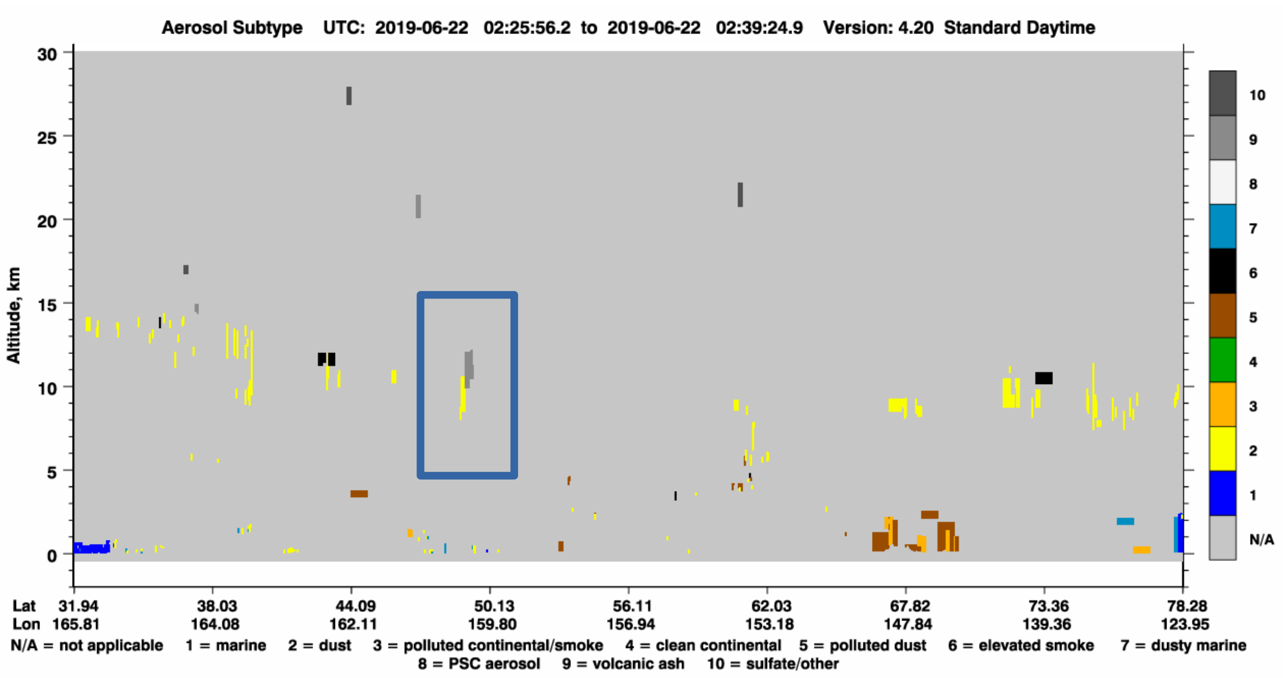
8. P9L240: The treatment of externally mixed ash and sulfuric acid would be more accurate through T-Matrix calculation than Mie Theory. I think this could be further discuss in the manuscript since it seems to be an important element.

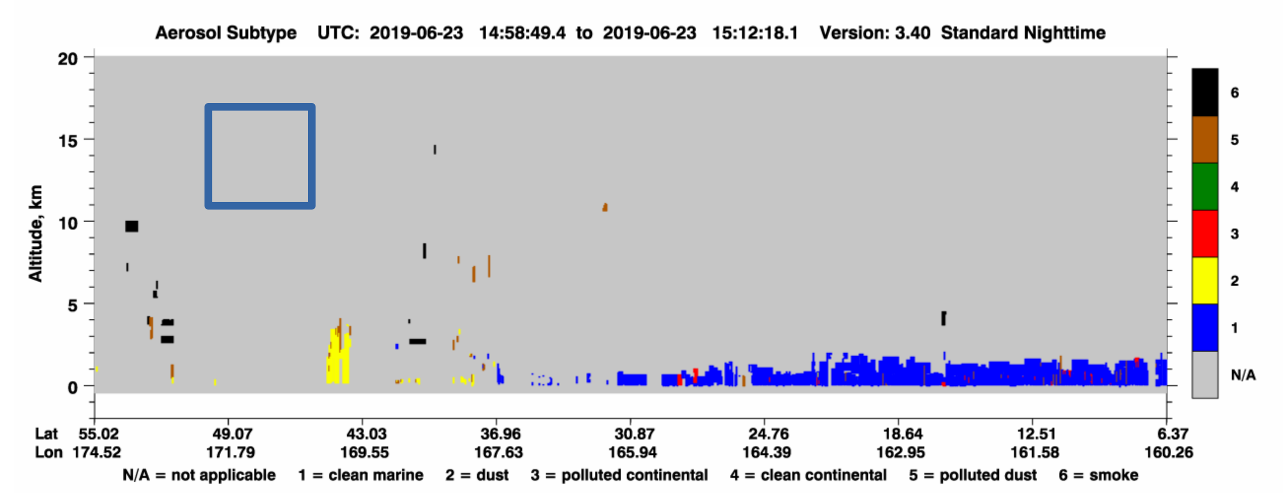
We agree with the reviewer that T-Matrix calculations are a powerful tool to determine the radiation interaction of non-spherical aerosols, such as volcanic ash. However, in this work in combination with the newly developed AERODYN module, we allow the formation of internally mixed particles, such as volcanic ash coated with a shell of sulfate and water. To our knowledge there is no T-Matrix code that can handle core-shell assumption. That is why we make use of Mie Theory assuming a core-shell mixing state.
Due to the coating, a spherical assumption for these mixed particles might be reasonable. It is only for consistency reasons, why we chose to apply the sphericity assumption also to the uncoated ash particles. Implementing coated non-spherical ash particles into ICON-ART or considering the non-sphericity of uncoated particles together with internally mixed ones remains the subject of future work.

9. P15L349: The other optical properties (depolarization/color ratio/vertical feature mask) from the plumes from CALIPSO are not shown. This would certainly help with the interpretation as well.

We agree that other optical properties retrieved from CALIOP measurements, such as depolarization ratio, give beneficial information about the composition of the aerosol cloud. In the current state of ICON-ART we don't have forward operators for these quantities. However, to our knowledge it is the first model that retrieves the total attenuated backscatter for internally mixed volcanic aerosol. This is why in this paper, we only compare the total attenuated backscatter at 532nm.

In the manuscript we argue that model results could help to interpret observations better. As an example, we took two images of the CALIOP Aerosol Subtype classification of two dates when the satellite passed over the volcanic cloud. In both images the blue rectangle highlights an area where the plume is located in our model result and also shows a signal in the total attenuated backscatter measured by CALIOP. For the first date, the detected aerosol (within the blue box) is classified only partly as volcanic ash. Based on our model result for the same date we would argue, that in fact the here classified dust is volcanic ash as well. For the other image, there is no aerosol type classified within the blue box. However, the total attenuated backscatter clearly shows a signal and our model results suggest that this is indeed volcanic ash.

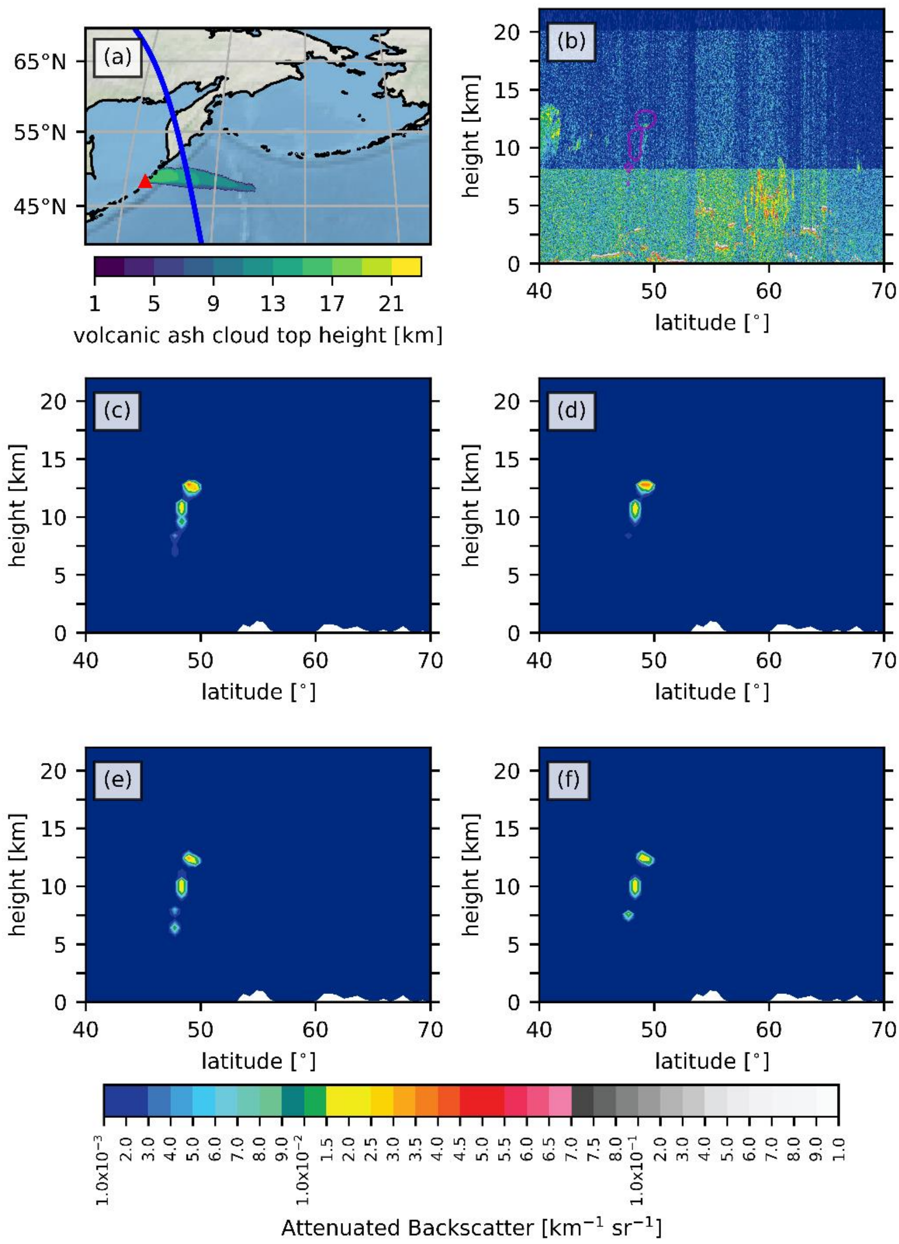




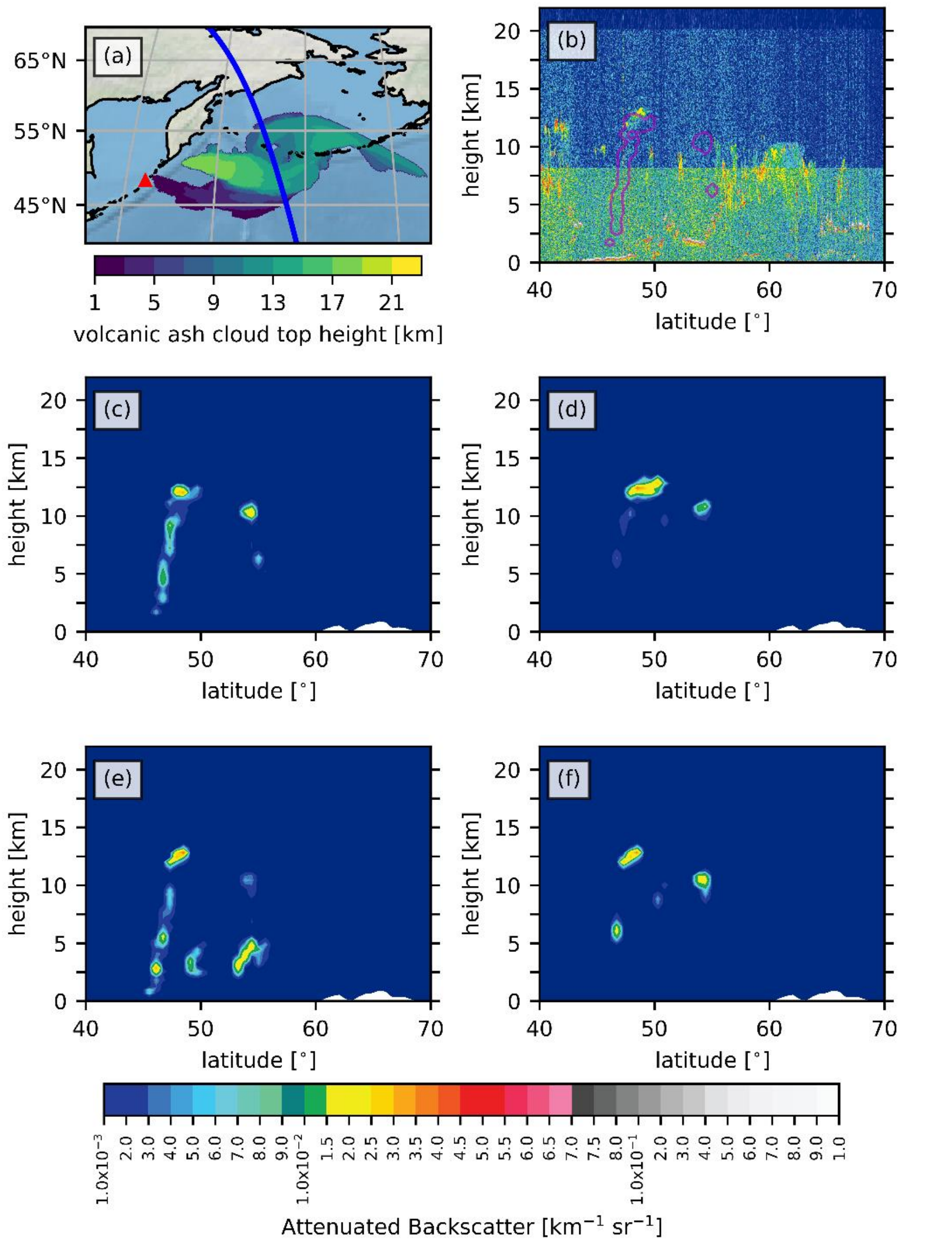
10. Figure 6: Does the model really do a better job representing the volcanic plume with the full dynamical-chemistry-radiation coupling? I'm not really certain that the figure demonstrate that since pieces of plume seen by the AERODYN-rad scenario do not appear clearly on the observations. See link to CALIPSO browse image crossing the volcanic cloud on Jun 22nd for additional obs. that could be used to validate model results. https://www-calipso.larc.nasa.gov/products/lidar/browse_images/show_v4_detail.php?s=production&v=V4-10&browse_date=2019-06-22&orbit_time=01-59-01&page=3&granule_name=CAL_LID_L1-Standard-V4-10.2019-06-22T01-59-01ZD.hdf

We evaluated four additional dates for which CALIPSO passes over the volcanic cloud of the Raikoke 2019 eruption. Furthermore, we extended the model comparison by the two no-rad simulation scenarios as requested by reviewer Arnau Folch. These plots will be displayed in the appendix of the manuscript.

The evaluation of these additional dates confirms our previous statement regarding the improvement of the forecast by including aerosol dynamics and radiation interactions. Only the very last date on June 25 shows no significant improvement.



2019-06-22 03:00 UTC
(a) CALIPSO ground track and modeled volcanic cloud top height
(b) Total Attenuated Backscatter at 532 nm measured by CALIOP
(c) AERODYN – rad
(d) no AERODYN – rad
(e) AERODYN – no rad
(f) no AERODYN – no rad



2019-06-23 02:00 UTC

(a) CALIPSO ground track and modeled volcanic cloud top height

(b) Total Attenuated Backscatter at 532 nm measured by CALIOP

(c) AERODYN – rad

(d) no AERODYN – rad

(e) AERODYN – no rad

(f) no AERODYN – no rad

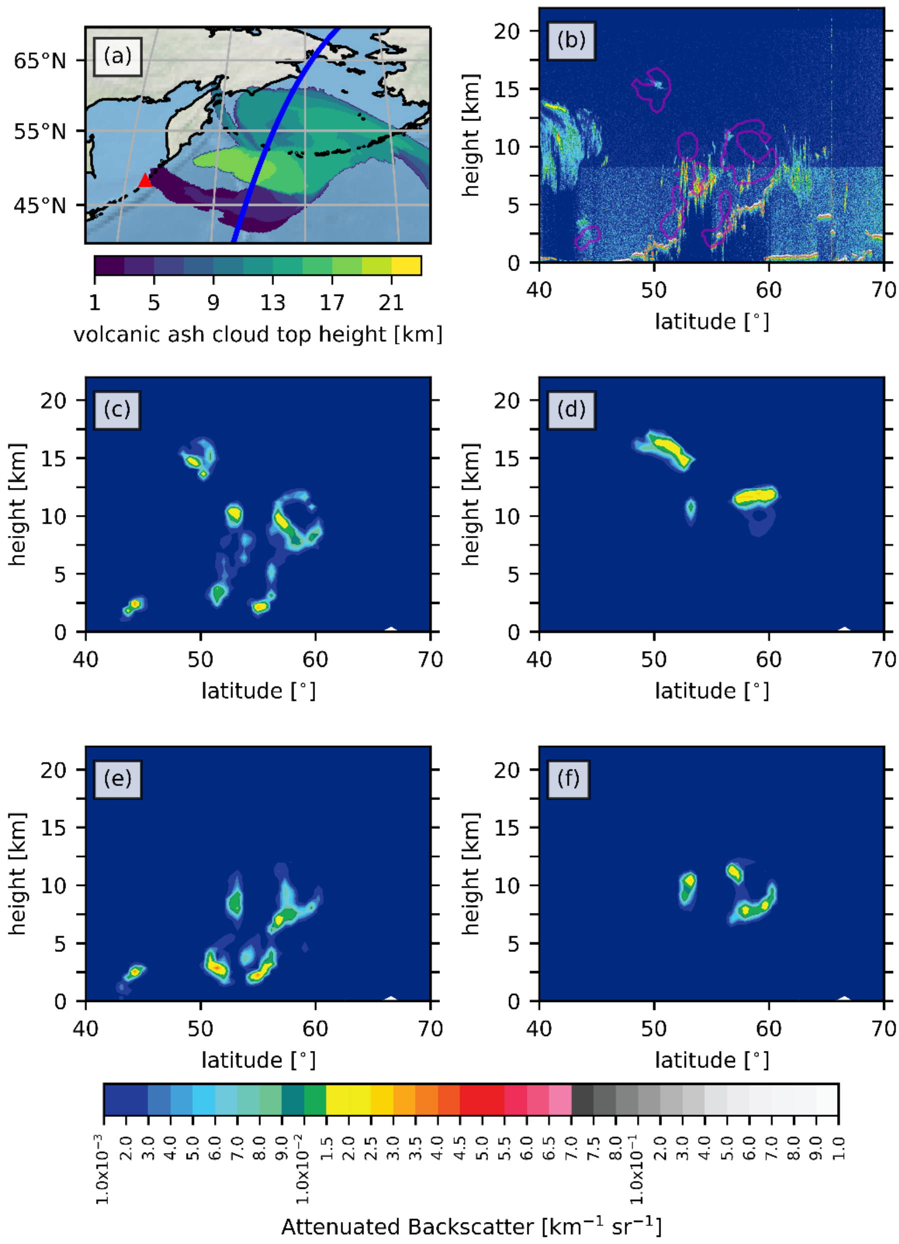
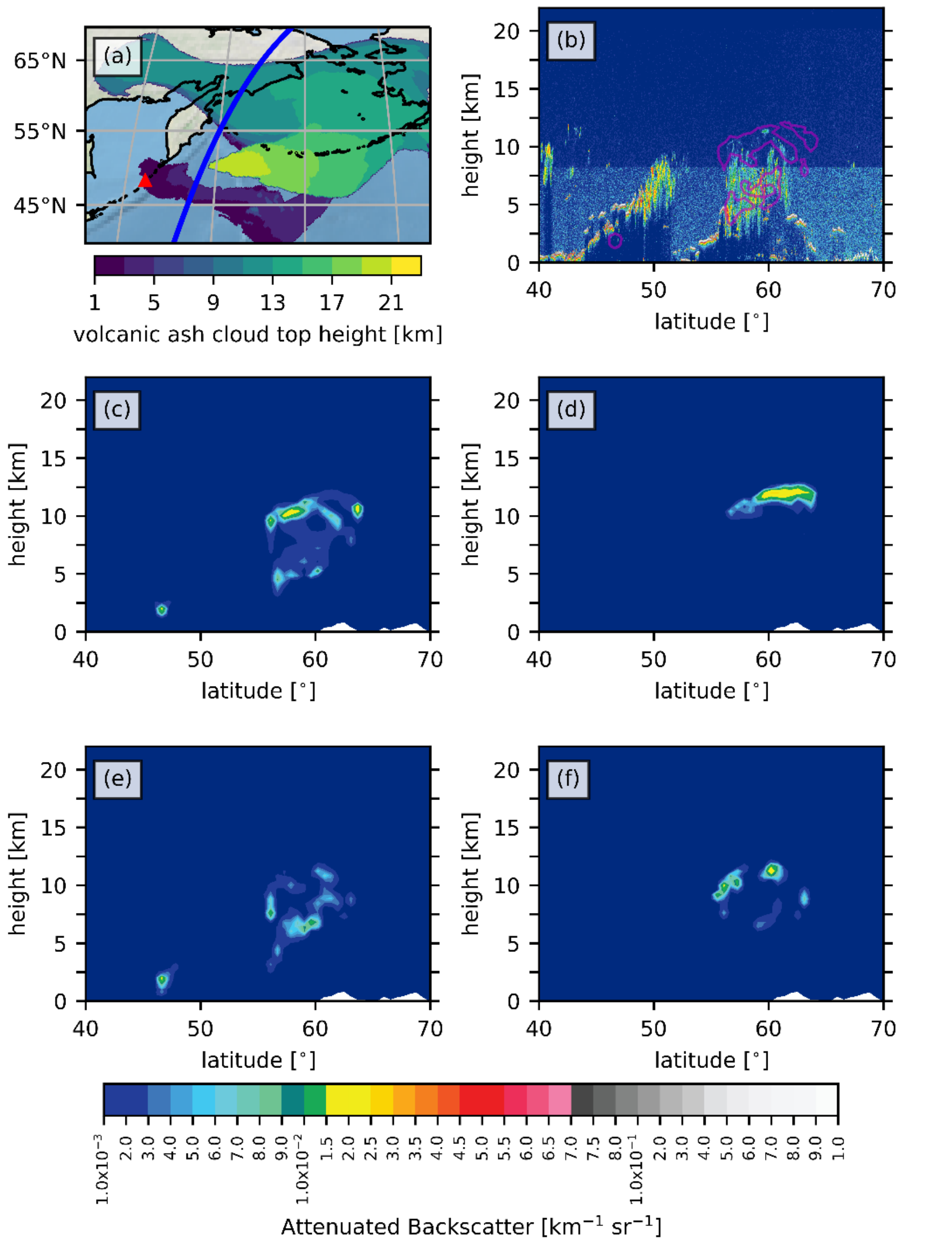
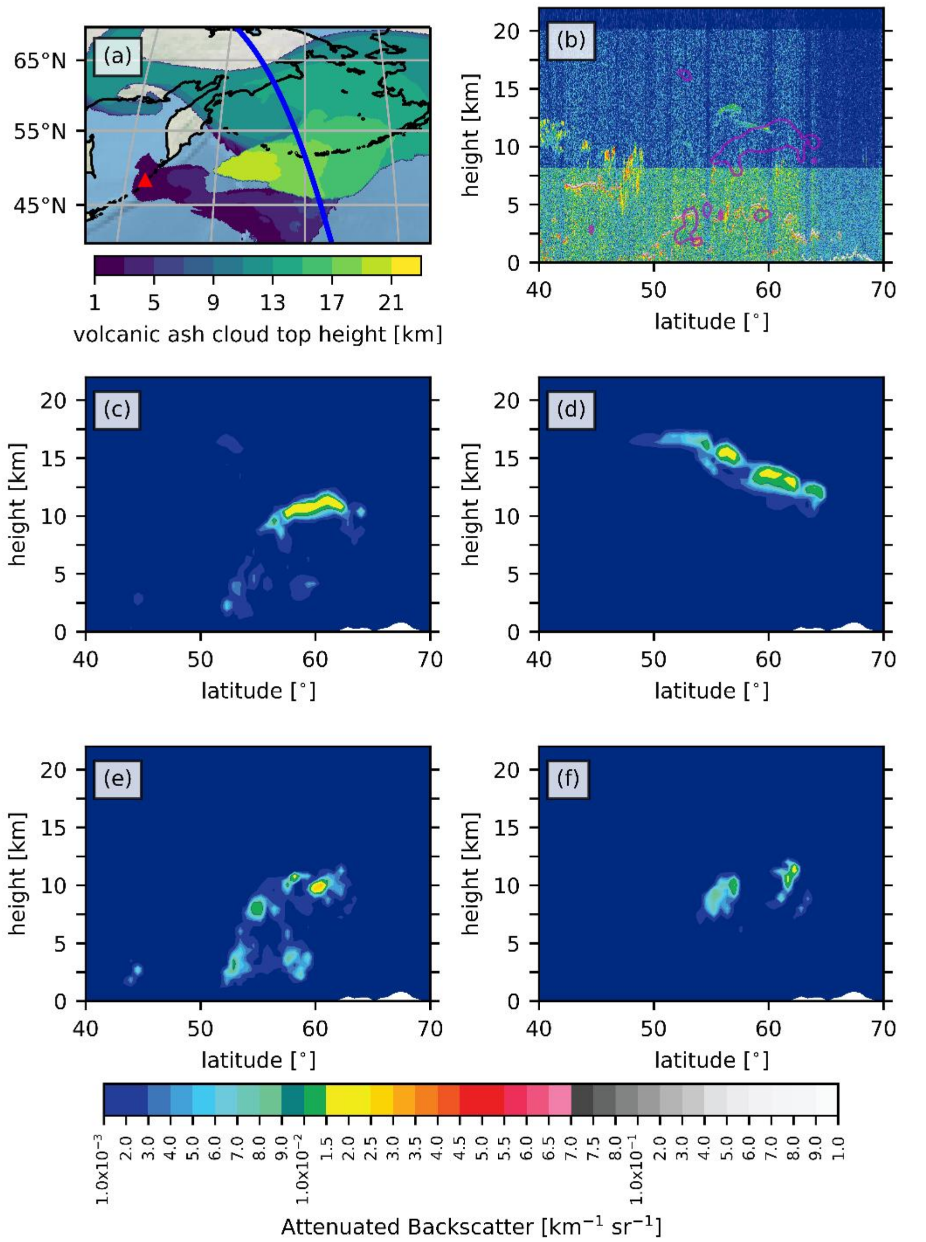


Figure 6. (a) CALIPSO ground track on 23 June 2019, around 15:00 UTC in blue color and location of Raikoke volcano as red triangle. The contour map shows the volcanic ash cloud top height for the AERODYN-rad scenario. (b) The CALIOP attenuated backscatter for 532 nm for the satellite position between 40° N and 70° N is displayed in the top right panel. The magenta line shows the 0.002 km^{-1}sr^{-1} contour of AERODYN-rad at 15:00 UTC. Middle and lower panels: Total attenuated backscatter for 532 nm of volcanic aerosols under the CALIPSO ground track on 23 June 2019, for the 15:00 UTC model output are displayed. (c) shows the result for AERODYN-rad, (d) for no_AERODYN-rad, (e) for AERODYN-no_rad, and (f) for no_AERODYN-no_rad, respectively.



2019-06-24 16:00 UTC
(a) CALIPSO ground track and modeled volcanic cloud top height
(b) Total Attenuated Backscatter at 532 nm measured by CALIOP
(c) AERODYN – rad
(d) no AERODYN – rad
(e) AERODYN – no rad
(f) no AERODYN – no rad



2019-06-25 01:00 UTC
(a) CALIPSO ground track and modeled volcanic cloud top height
(b) Total Attenuated Backscatter at 532 nm measured by CALIOP
(c) AERODYN – rad
(d) no AERODYN – rad
(e) AERODYN – no rad
(f) no AERODYN – no rad

11. Figure 7: Even if the model indeed do a better job by including the dynamics and radiation to remove ash, it does not capture well small-scale variations. Could you further explain why it's not the case? Maybe incorporating more accurate source terms based on HIMAWARI-8 would help with that.

This is a very good point. Although the overall agreement is very good, there are small-scale variations in the AHI retrieval that don't have a corresponding model result.
At this point we should distinguish between two periods, the eruption period, roughly during the first 12h, and the quiet period during which the volcano did not emit ash anymore.
In this study we emit volcanic ash with a constant emission rate over 9h. We know from satellite images (GEOS17, Himawari-8) that Raikoke emitted ash with several longer and shorter puffs. The last, rather short puff happened on 22 July 2019 at around 07:10 UTC. This explains well the offset between model result and observation, and also the small-scale variations in the observation during the eruption phase. Characterizing these puffs in terms of height and mass eruption rate (and thus time dependent eruption rate) is the topic of ongoing work.
After the eruption has stopped, the small-scale variations in the AHI retrieval are due to deficiencies or limitations of the retrieval algorithm, as any increase of measured ash cannot be associated with an emission.
In order to make this clearer in the manuscript, we add additional information to l.368.

As Raikoke did not erupt continuously over these 9 h, the offset between simulation and observation as well as the small-scale variations in the observation during this period can be explained.

Furthermore, we add one and rephrase one sentence in l. 373:
The small-scale variations in the observation might be due to deficiencies or limitations of the retrieval algorithm, as no new ash is emitted during this period. We can see a very similar decay and stabilization of ash mass for the AERODYN-rad scenario in green.

12. P17L375: It would be interesting to know which processes contribute to the removal of ash in the model. I believe the growth term that lead to the removal by sedimentation, what about ash-ice interaction and wet deposition?

In ICON-ART we account for sedimentation, dry deposition and wet deposition (scavenging by raindrops below clouds). These processes are active for all aerosols for all presented simulation cases, i.e. they are not exclusively linked to the AERODYN development. However, the presented setup of ICON-ART does not account for ash-ice interaction or CCN activation of aerosol yet. Combining aerosol aging with aerosol activation will be subject of future development.
In order to have this information in the manuscript as well, we add the following in l.184:

The removal of aerosols from the atmosphere is modeled by three different processes: sedimentation, dry deposition and wet deposition. In ICON-ART wet deposition describes scavenging by raindrops below clouds.

Furthermore, we discuss the differences of these three mechanisms in l. 377 ff.:
Additionally, we would like to note that the prevailing settling mechanism of aerosol after the Raikoke 2019 eruption for all our simulation scenarios is due to sedimentation. Dry

deposition is only relevant for aerosol near the ground. Wet deposition should also play a minor role during the first days after the eruption, as most of the volcanic ash is emitted above cloud level.

13. Figure 9: More data are needed to verify the model outputs. e.g. CALIPSO and OMPS.

As already discussed in our answer to comment 7, we were not able to retrieve additional meaningful OMPS-LP data for plume top heights.
In contrast, there exist several CALIPSO overpasses (which we show in our answer to comment 10). The measurements of total attenuated backscatter at 532 nm on these dates show a signal which can be associated with volcanic aerosol. However, in the scope of this work we were not able to define an objective quantity that allows us determining the volcanic cloud top height in CALIOP measurements. That is the reason why we constrain the comparison between CALIOP and ICON-ART to the more qualitative Fig. 6.

14. P20L431: I believe that measurement uncertainty from OMPS could be better addressed. The vertical resolution of the instrument is probably near 1-2 km. Could you add the corresponding error bar in figure 8. In addition, I'm pretty confident that additional information on volcanic cloud top height could be found by analyzing additional OMPS data.

The vertical sampling of OMPS-LP is 1km which gives us +/- 0.5km accuracy in the peak attribution. The remaining uncertainty in the pointing is about 0.2km. The latter is rather systematic. The aerosol retrieval has a vertical resolution of about 3km which smears the peak, however, won't displace it. This is why we estimate the measurement uncertainty with +/- 0.7km.
As suggested, we add an error bar to the OMPS-LP measurement in figure 8.

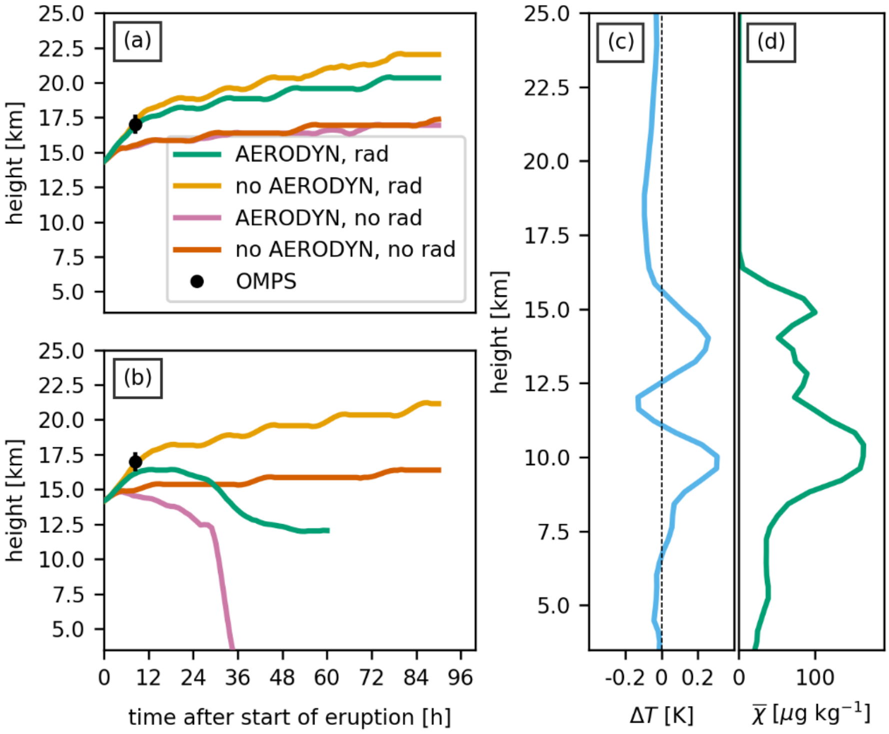


Figure 8. (a) and (b) Evolution of height of volcanic ash cloud top after the onset of the eruption on 21 June 2019, at 18:00 UTC. The yellow curve represents the no_AERODYN-rad scenario, the green curve AERODYN-rad, the pink one AERODYN-no_rad, and the orange one represents the no_AERODYN-no_rad scenario. Panel (a) shows the ash cloud top of particles in the accumulation mode, (b) of particles in the coarse mode, respectively. The black circle depicts the volcanic cloud top height obtained from OMPS-LP. (c) Mean temperature difference (AERODYN-rad – AERODYN-no_rad) in volcanic ash cloud columns on 23 June 2019, 12:00 UTC. (d) Mean volcanic ash concentration $\bar{\chi}$ for the same model columns as in (c) for AERODYN-rad.

Additionally, we add in l.161:

Due to uncertainties in pointing and vertical sampling we estimate the measurement error with +/- 0.7km.