Atmospheric Chemistry and Physics Discussions

Atmos. Chem. Phys. Discuss., referee comment RC2
https://doi.org/10.5194/acp-2021-471-RC2, 2021

Comment on acp-2021-471

Anonymous Referee #2

Referee comment on "Aerosol responses to precipitation along North American air trajectories arriving at Bermuda" by Hossein Dadashazar et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-471-RC2, 2021

Hossein Dadashazar et al. investigated the impact of precipitation on aerosol particles along single particle back trajectories to Fort Prospect measurement station located on Bermuda. An assessment of the seasonal transport climatology to this station is provided by clustering the trajectories into different transport corridors. An investigation of the sensitivity of PM2.5 to experienced precipitation during transport is performed. This showed that air-masses arriving from North America during wintertime experienced the highest accumulated precipitation, which was found to correspond with large reductions in the PM2.5/ΔCO. These results are complemented and discussed with respect to GEOS-Chem model sensitivity simulations that provide insight into the role of large-scale precipitation scavenging versus convective wet scavenging, and a recent case study from a research flight from the ACTIVATE aircraft measurement campaign.

Overall, the manuscript is very well written and structured. There are some minor instances where the readability can be improved, which I have indicated below. The scientific methods used in this study are accurate, although I do have some reservations about the applicability of single particle trajectories to assess transport during the ACTIVATE flight measurements.

This study is well suited to the scope of ACP and I do recommend this study for publication.

I have listed some questions and suggestions below to address prior to publication.

General comments :-

1.) The underlying results presented in this interesting study are based around combining in-situ observations with single particle trajectories calculated from the HYSPLIT trajectory model driven by GDAS reanalysis data. As with any study that is based around a model it is important to acknowledge any uncertainty in the results that are attributed to the modelling framework. Demonstration of the role of uncertainty in (a) the single-particle trajectory calculations and (b) the choice of reanalysis data, and their implications on the conclusions presented is currently missing and requires further attention.

(a) Uncertainties in the representation of air-mass history associated with single particle trajectories can be large. The accuracy of a single particle trajectory in representing the transport history compared to more detailed, e.g. dispersion modelling will depend on the prevalent atmospheric conditions and the duration of the trajectories e.g. https://doi.org/10.5194/acp-9-8857-2009. Accordingly single-particle trajectory analysis is typically applied to investigate the average transport climatology to a receptor station by averaging many thousands of trajectories over long-time periods e.g. https://doi.org/10.5194/acp-13-3643-2013, and this approach is also applied in the majority of this study. However, for the comparison against the aircraft observations over a short-time period single-particle trajectories are also applied which will involve large uncertainties. As a minimum it would be appropriate for this section to run HYSPLIT in its trajectory ensemble configuration. The divergence of those trajectories would provide a more quantitative estimate of the uncertainty associated with the single particle trajectories currently shown.

(b) There are numerous trajectory models available aside from HYSPLIT (e.g. Flexpart). For the calculation of single particle trajectories from these models I would expect the largest differences in the resulting trajectories to be due to the choice of meteorological (reanalysis) data used as input, e.g. https://doi.org/10.1080/10473289.2005.10464758; https://doi.org/10.5194/asr-2-65-2008. Currently there is no discussion of this uncertainty. The study uses GDAS reanalysis data to drive HYSPLIT. HYSPLIT can be run using other reanalysis products, e.g. ERA-Interim, ERA5. How would the results change if a different reanalysis product (e.g ERA-Interim) was used, with respect to the air-mass history and experienced precipitation during transport? Significant differences in precipitation exist between different reanalysis products, e.g. https://doi.org/10.1002/2017RG000574; https://doi.org/10.1016/j.jhydrol.2020.124632. How would the results change if you collocated precipitation from the Global Precipitation Climatology Project (GPCP) along your GDAS trajectories?

2.) In the study the surface in-situ measurements are "averaged every 6 hours to match the time frequency of the trajectory analysis". It is unclear why this is required and some clarification is required as to why this approach was employed. With this type of trajectory modelling (single particle trajectory calculations) there are no major limitations associated with computational cost or data storage (very fast model, trajectory output data small). To my understanding the current approach will result in the collocation of in-situ data averaged over 6 hours, to a trajectory arriving at an instantaneous time. Are the measurements averaged such that the single trajectory arrives in the middle of the 6hour time window? Why are hourly trajectories not calculated, and collocated to the hourly measurements, as has been performed in previous similar studies, e.g. https://doi.org/10.5194/acp-13-3643-2013. This would be advantageous as it would

significantly improve your statistical analysis (providing 24 trajectories per day as opposed to the current 4 trajectories per day), and more confidence that the trajectories represented the air-mass history associated with the hourly in-situ measurements.

3.) The study employs Modern-Era Retrospective analysis for Research and Applications-Version 2 (MERRA-2) as a data source for some aerosol parameters that are not measured in-situ at the station. How was this data connected to the HYSPLIT trajectories? The methodology (line 160) does not state whether this data was collocated along each of the individual trajectories prior to averaging. This would be beneficial; as it would also allow for a comparison of the reanalysis data to the in-situ measurements for certain key parameters to quantify the uncertainty associated with MERRA-2 as well as demonstrating how key parameters from MERRA-2 varied during transport, which would aid in understanding the role of precipitation on aerosol lifecycle during transport.

4.) As the key parameter used in the study is the accumulated precipitation along trajectories (APT) can the authors please provide some more details on how this was calculated/processed within the paper? Specifically, what does the precipitation diagnostic obtained from HYSPLIT-trajectories represent? Does it represent a precipitation rate that was converted to a total amount at each hour along the trajectory and then summed? Does it represent the precipitation at the height of each trajectory point, summed along the entire trajectory, or a column total precipitation at each point along a trajectory. If the latter, how did the authors process this data to ensure that the APT represents below cloud removal of aerosol? Some clarification is required here as if an individual trajectory is not at the height where the precipitation is occurring then this should be accounted for in the subsequent data processing.

5.) The study focusses on separating out transport corridors using a clustering method, however, the role of variation in vertical transport is not discussed. In figure 4 (b) what is the vertical transport climatology between cluster 1 and 2. What percentage of these clusters are dominated by low level transport more in which wet scavenging is more likely to play a role on aerosol removal? This should be considered in the analysis, and discussed. One solution would be to cluster the trajectories associated with cluster 1 and 2 with respect to height above the surface during transport. Furthermore, whilst the focus of the study is on the role of precipitation on aerosol during transport, the role of in-cloud scavenging is not discussed. What is the relative importance of in-cloud removal of aerosols during transport? This could be assessed by calculating the time spent in-cloud / out of cloud during transport to Bermuda. Such further analysis is required in order to support the final statement "wet scavenging processes in models require stronger constraints than other aerosol microphysical/chemical processes to improve the forecasting". Regardless, I recommend that this statement is rephrased slightly, to e.g. "it is important to strongly constrain we scavenging processes in models to improve the forecasting over the WNAO".

6.) Key references that have performed similar analysis on the role of precipitation on the aerosol lifecycle are currently missing e.g. https://doi.org/10.5194/acp-13-3643-2013. Whilst performed in a different study region, such studies deserve inclusion in the discussion with respect to, for example, the interplay between precipitation and aerosol nucleation during the aerosol lifecycle.

Minor comments :-

The study region is defined as a "ideal natural laboratory" within the same sentence that it is highlighted that it is a region that the region experiences pollution, i.e. not natural aerosol sources. A slightly rephrasing may be considered here.

Line 41: salt > sea salt

Line 65: "(Sorooshian et al., 2020), especially" > is the especially required in this context, simply remove?

Line 76: "along those trajectories": trajectories mentioned for the first time here in the introduction. Suggest introducing first, for example when first mention air-mass history can explain that this can be calculated using trajectory models.

Line 77: Would benefit from some detail explaining why wet scavenging rates are difficult to constrain over the WNAO compared to other regions.

Line 82: "rain along trajectory pathways" > consider revising, e.g. precipitation transport history prior to arrival.

Line 83: Would benefit from an appropriate reference.

Table 1: This table could be improved in places. The spatial resolution of certain datasets is missing. Is "Back-trajectory" a parameter? Consider splitting the table to provide one for in-situ observations, and one for model data.

Section 2.2: This section outlines the reanalysis data used within the study but does not mention the reanalysis data used in the trajectory modelling (GDAS).

Line 160: “converted to 6-hour data”. Can you please clarify what is meant by converted? Averaged, or instantaneous fields?

Line 165: “ending altitude of 100 m (AGL)”. The height of the instrumentation AGL at the station is not provided for comparison. Due to the native resolution of the GDAS reanalysis data used for the trajectory calculations care needs to be taken for high altitude stations, as the average trajectory height at a location calculated by the trajectory model from 1-degree resolution reanalysis data may differ from the actual height.

Line 171: “with the “model vertical velocity” method”. Please consider rephrasing for clarity. I believe that this means that the vertical velocity is obtained from the underlying reanalysis data rather than being calculated from the HYSPLIT “model”, see https://www.arl.noaa.gov/documents/workshop/NAQC2007/HTML_Docs/trajmetd.html

Line 175: “remaining sections of the paper are based on 4-day (96 hr) back-trajectories.” I assume these are not from different trajectory calculations, but are the same trajectories simply truncated to end at 96hr instead of 240hr. Please clarify.

Line 185: “A weight function following the method of Dimitriou et al. (2015) was applied”. Please explain this method, demonstrating the equations used, for clarity.

Line 224: ^{222}Rn > define acronym - Radon 222 (^{222}Rn)

Line 269: “It can be deduced from Fig. 1 that based on the farther reaching source areas”. The average wind speed can be easily calculated from the data in the trajectories, so there is no need to deduce from the figure. Please calculate instead.

Line 289: “Fort Prospect data to” > “data from Fort Prospect station to”

Figures 2/3: One colour bar is sufficient for all subplots if the limits are constant for each.

Line 317: Consider rephrasing to improve readability “including when resolved by season”

Line 333: It is stated that 0.5 degree grids are used, however, to my understanding the GDAS data used is 1 degree. Why is the data averaged over a higher resolution than the underlying data source?

Figure 4: Please provide clarification on how Figure 4 (b) was created. On line 365 it states "Using only two clusters increases" which indicates that you have performed a new clustering, this time setting the number of clusters to 2 when you cluster. Or, does this mean you have simply grouped the clusters in Fig. 4a into two groups?

Figure 4 (b). For clarity, would benefit from showing the trajectory members associated with each cluster in a different colour, e.g. light grey. This would help show the benefits of sub-sampling the data using clustering over simply sectoring the transport pathways (e.g. select only trajectories that spend 95% time within certain lat/lon sectors).

Line 382: Please consider rephrasing "more ideal" to improve readability.

Line 382: "identifying potential wet scavenging effects". Some clarification is required on how it was determined that cluster 1 was more "ideal". Why was clustering of trajectories chosen as a method rather than sectoring? For example, an alternative strategy would be to bin the in-situ data by experienced precipitation during transport to identify the impact of wet removal for different transport corridors. This would provide valuable insight into the role of precipitation on the aerosol lifecycle for different aerosol regimes for this region.

Line 403:405: This sentence is quite long and a little hard to digest. Please consider rephrasing to improve readability.

Line 459: "four bins of APT chosen in such a way to provide similar numbers of data points per bin". Some clarification is needed here. What are the bin limits for each bin? Also, why is this approach required? Can the authors please demonstrate how the sensitivity in Figure 7 changes if a constant bin width is used, as performed in previous similar studies, for example Fig. 15: https://doi.org/10.5194/acp-13-3643-2013. A histogram showing the counts per bin can be provided, and a minimum number of counts for a bin to be considered can be implemented in such an approach. Statistics can easily be improved by using hourly trajectories (see general comment 2).

Line 506: "This can be explained by how the high APT days" > consider rewording, e.g. "how days experiencing high APT…"

Line 587: "2017 are used for analysis, which is a representative year". The authors should demonstrate in the SI, how it was ascertained that 2017 is a representative year by, for example, showing how the average precipitation/meteorology for this year compared to the average during 2015-2019.

Line 552: Could the authors please clarify how the mode statistics were calculated. I believe the correct procedure to obtain the average modal parameters provided in the table would be to calculate the modal parameters for each individual VSD, and then average these to obtain the average modal parameters, rather than calculating average modal parameters from the average VSD. Which approach was used?

Line 608:611: Could the authors please add some caveat to make it clear that these results pertain to a model for which the representation of aerosol – cloud – interactions are highly uncertain, and therefore, do not necessarily represent accurately what is happening in nature.

Line 618: "this study to unite the greatest potential for wet scavenging" please consider rephrasing to improve readability.

Line 658: "As a successful validation of the technique, no rain accumulated…" please consider rephrasing to improve readability.

Line 656: "Four HYSPLIT back-trajectories are shown (Fig. 10a)". Please consider making this plot larger and giving it its own subplot label.

Line 658: Sentence beginning "As a successful validation …" please consider rephrasing to improve readability.

Page 23: Consider placing key statistics from the ACTIVATE research flight into a table to improve accessibility.

Line 729: "surface on days simultaneous with high APT trajectories." please consider rephrasing to improve readability.

Line 731: "in contrast to less change" > "to a smaller change in.."