

## Response to Revisor 2

### Overall comments to the revisor

The authors would like to express their gratitude for the comments about the manuscript. Below you will find a detailed answer to all comments and how they were addressed in the text. We reproduce Revisor's comments and add our responses in *red italicized font*. In addition to those points, the authors would like to highlight that the changes throughout the text are in the uploaded marked-up version of the manuscript.

### Answers to the major comments:

**1) Physical Contradiction and the Twomey Effect Mechanism:** a fundamental concern exists regarding the application of the LWC - reff relationships derived in Section 2.5. The Twomey effect is physically defined as an increase in cloud albedo and optical depth due to an increase in droplet number concentration (Nd) and therefore a decrease in reff for a constant liquid water content (LWC). The Problem of Fixed Nd: By deriving and utilizing fitted relationships between LWC and reff for specific scenarios (Clean vs. Polluted), the authors effectively assume a fixed Nd for a given scenario. Contradiction in Forcing Drivers: If the IRF within a given scenario is driven by a change in reff while Nd remains fixed, it contradicts the Twomey effect. Specifically, if a negative IRF (larger albedo) is attributed to a larger reff under these fixed-fit conditions, the physics would be inverted. The authors must verify that a negative IRF corresponds to a reduction in reff as aerosol loading increases.

*R: The Problem of Fixed Nd -We appreciate the reviewer's observation, as it made us realize that the text is inaccurate and needs further development. An analysis of the different flights used revealed that warm clouds with different characteristics were traversed, but formed in scenarios characteristic of aerosol availability, confirming that a constant Nd was not considered. Furthermore, Nd and LWC measurements were performed using independent instruments (WCM-2000 and FCDP, respectively), following the same methodology used by Reid et al. (1999), i.e., adjustments to the relationships between LWC and reff for the different identified scenarios. The fact that Nd is not constant can also be seen in Figures 1 and 2 below. These figures show that, for a given fixed bin of LWC, the Nd and reff values vary differently between the IOPs, confirming that the microphysical mechanism of the Twomey effect occurs. For the range between 0.2 and 0.4 g/m<sup>3</sup> in the LWC, in particular, higher Nd values and lower reff values occur in IOP 2 than in IOP 1, as expected. This discussion, including Figures 1 and 2 (Figure 15 in the revised version), was included in section 4 of the revised version. Contradiction in Forcing Drivers - Both Figure 8 (corresponding to the first clean atmosphere reference, considering constant irradiance) and Figure 13 (second reference, with irradiance varying seasonally) present in the manuscript and reproduced below show that the medians of IRF<sub>daily</sub> are negative for both years. When these medians are evaluated together with the seasonal evolution of the reff medians for 2014 (equal to 14.3, 10.4*

and 5.6  $\mu\text{m}$  for the clean, transition1 and polluted1 scenarios, respectively) and for 2015 (14.2, 9.5 and 5.5  $\mu\text{m}$  for the clean, transition1 and polluted scenarios, respectively), it is possible to see that the IRFdaily distributions are shifted towards negative values while the reff distributions are shifted towards lower values, with the lower reff values occurring due to an increase in aerosol load. This discussion was also included in section 4 of the revised version of the manuscript.

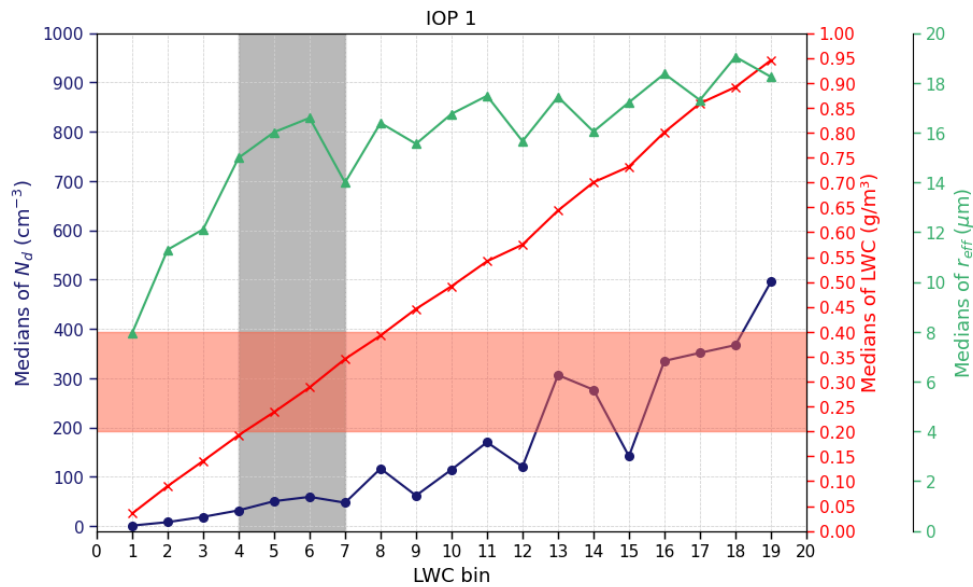


Figure 1: Median's variations ( $N_d$ , LWC e reff) along LWC bins for IOP1. Bins 4 to 7 correspond to the LWC range (0.2 to 0.4  $\text{g/m}^3$ ) considered for filtering the clouds analyzed in the study.

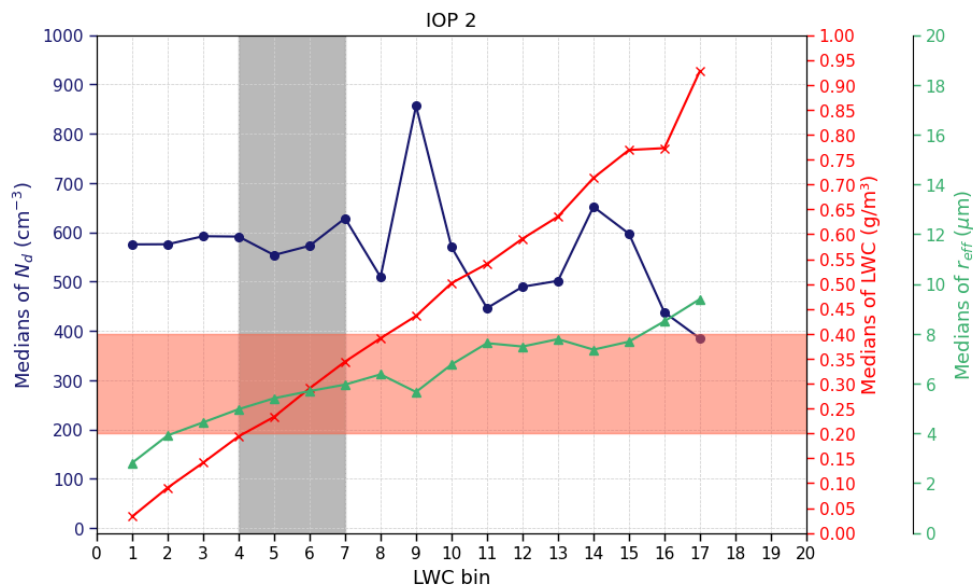


Figure 2: Median's variations along LWC bins for IOP2. Bins 4 to 7 correspond to the LWC range (0.2 to 0.4  $\text{g/m}^3$ ) considered for filtering the clouds analyzed in the study.

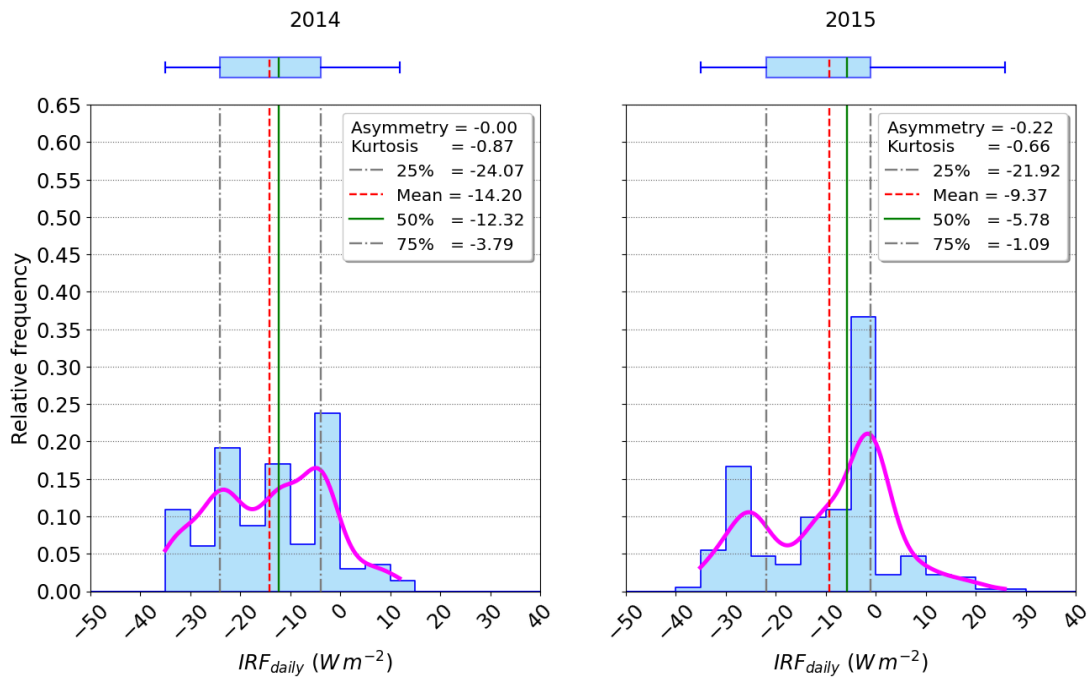


Figure 3: Reproduction of manuscript's Figure 8, showing boxplots and histograms of  $IRF_{daily}$  for 2014 and 2015 for  $fc = 100\%$  according to the first reference for a clean atmosphere.

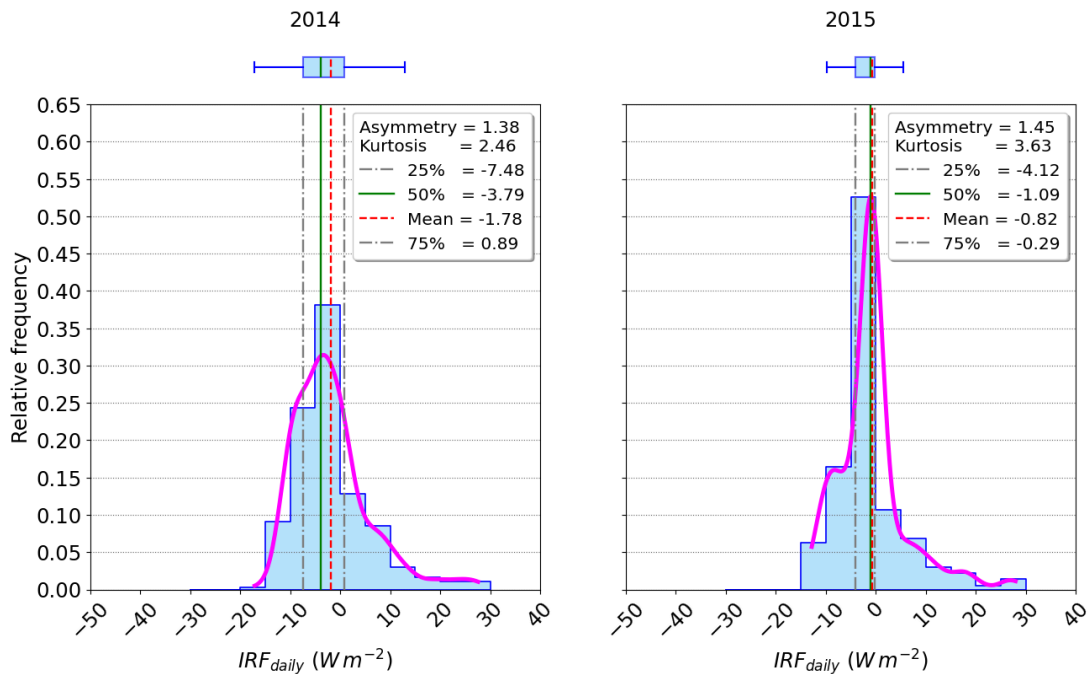


Figure 4: Reproduction of manuscript's Figure 13, showing boxplots and histograms of  $IRF_{daily}$  for 2014 and 2015 for  $fc = 100\%$  according to the second reference for a clean atmosphere.

**2) Methodological Validity of Atmospheric Reference States:** The study relies on two distinct "clean" reference states to calculate IRFaci (Eq. 10). Fixed Reference Issues: The use of a fixed irradiance value ( $689.9 \text{ W m}^{-2}$ ) derived from the campaign's 19 cleanest days is problematic. Radiative forcing variations under a fixed reference are likely dominated by natural shifts in cloud macro-properties (height, fraction, LWP) driven by thermodynamic and dynamic environments rather than aerosol effects. The conclusion that these results serve as a benchmark is weakened by this seasonal contamination. Representativeness of HALO Data: The seasonal reference approach utilizes only two HALO flights (AC09 and AC18) to define the background for the entire polluted dry season. It is unclear if these two specific days are statistically representative of the broader seasonal variability in a region as dynamic as the Amazon. A sensitivity analysis or justification for the representativeness of these flights is required.

*R: Fixed Reference Issues - From this comment, we were able to verify the need to include more details about the gas and temperature profiles used in all simulations performed by LibRadtran. Therefore, we clarify that the vertical profiles of  $\text{O}_2$ ,  $\text{O}_3$ ,  $\text{NO}_2$ ,  $\text{CO}_2$ , water vapor, air density, temperature, and pressure (Figures 5 and 6 below) were provided by the "standard tropical atmosphere" file (afglt.dat) made available by LibRadtran. For the profiles of more rarefied components (Figure 7), in turn, representative tropical atmosphere data provided by the Earth Observation Data Group (EODG) of the physics department at the University of Oxford (<http://eodg.atm.ox.ac.uk/RFM/atm/minor.atm>) were used. All the profiles mentioned were used equally in all simulations, seeking to follow the definition of "instantaneous" forcing, namely, forcings obtained with the constant T tropospheric and stratospheric profiles. This observation was included in section 2.7. The use of a variable/seasonal reference based on aerosol availability, while also keeping the previous profiles fixed, aimed precisely to reduce the seasonal contamination of other variables and isolate the effect of aerosols alone on indirect forcing, since the effects of seasonal contamination due to dynamics on macrophysical characteristics of clouds are embedded in the distributions of these same properties (LWP, CBH, CTH). More detailed discussions on this point were included in sections 2.8 and 4. Representativeness of HALO Data - flights AC09 and AC18 were conducted over two regions practically unaffected by biomass burning plumes from the deforestation arc, characterizing the cleanest possible atmosphere among all flights performed, as indicated by the average values of numerical aerosol concentration ( $N_a$ ) and CCN in Table 1. Despite the small number of samples (two cases), these flights are representative of the cleanest possible atmospheric regime within the polluted season in the region, which is why they constitute the best available samples to characterize the second clean scenario used in the study. This discussion was presented in subtopic 2.3 of the new version of the manuscript.*

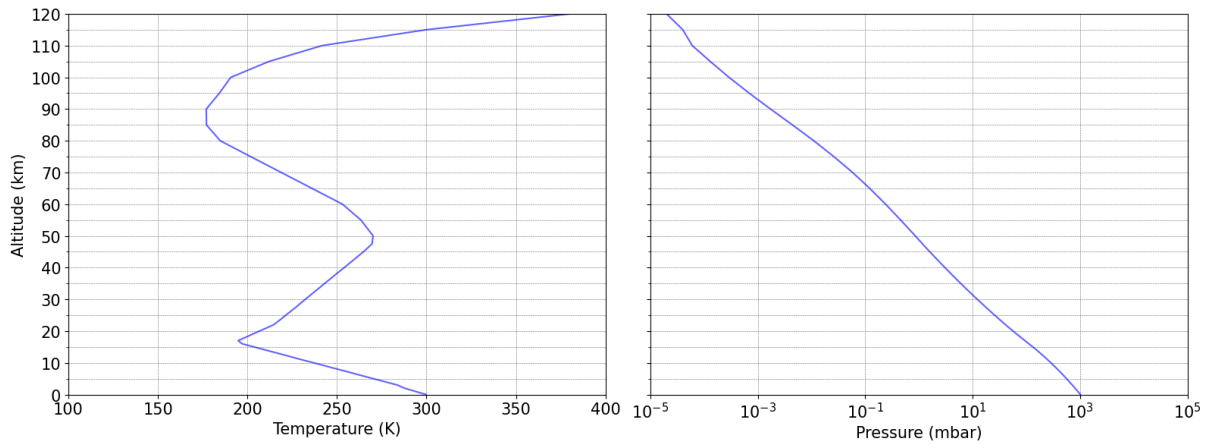


Figure 5: Vertical profiles of Temperature and Pressure from afglt.dat

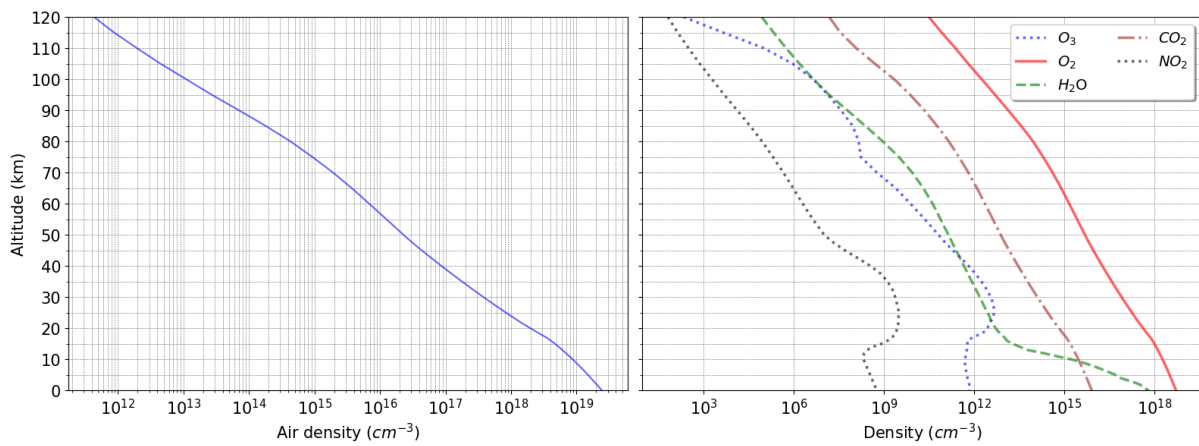


Figure 6: Vertical profiles of gases provided in the afglt.dat archive.

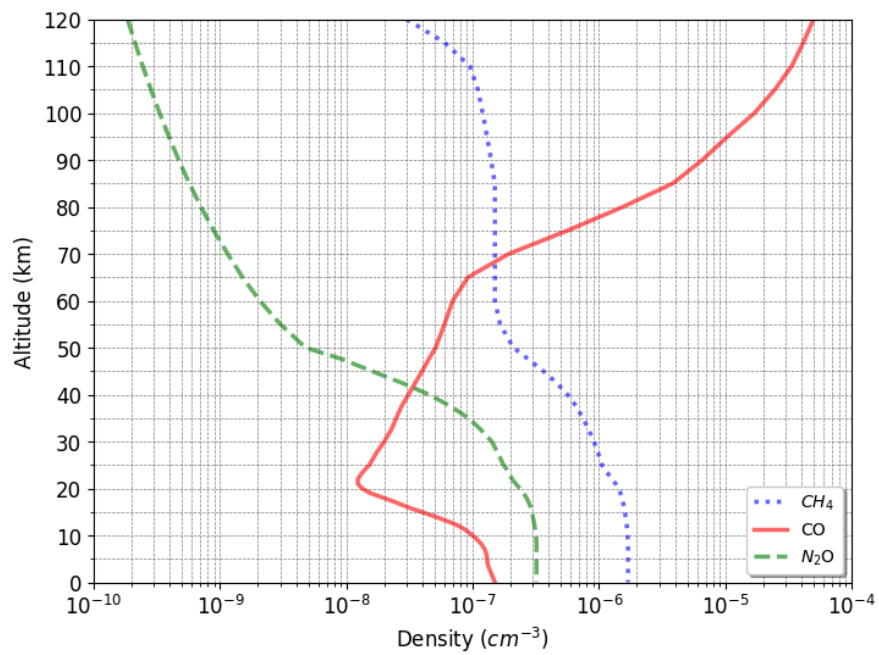


Figure 7: Vertical profiles of additional gases. Provided by EODG, Oxford.

*Table 1: Average concentrations of  $N_a$  and CCN measured during some HALO flights during 2014. Adapted from CECCHINI et al., 2017.*

<i>Flight</i>	<i><math>N_a</math> (<math>cm^{-3}</math>)</i>	<i>CCN (<math>cm^{-3}</math>)</i>
<i>AC18</i>	<i>744</i>	<i>408</i>
<i>AC09</i>	<i>821</i>	<i>372</i>
<i>AC07</i>	<i>2498</i>	<i>1579</i>
<i>AC11</i>	<i>2691</i>	<i>1297</i>
<i>AC12</i>	<i>3057</i>	<i>2017</i>
<i>AC13</i>	<i>4093</i>	<i>2263</i>

**3) Consistency of Aircraft Sampling and Microphysics:** The derivation of  $re_{ff}$  from flight medians across IOP 1 and IOP 2 (Fig. 3) requires more context regarding the sampling strategy. Flight Patterns: The authors must clarify if the flight patterns for the G-1 during IOP 1 (wet) and IOP 2 (dry) were consistent. Discrepancies in sampling altitudes or cloud types (cumulus vs. stratocumulus) between the two periods could introduce significant biases in the LWC -  $re_{ff}$  fits. Vertical Profiling: It is unclear if the  $re_{ff}$  values represent layer averages or specific flight levels. Since  $re_{ff}$  typically increases vertically from the cloud base, the "middle layer" median approach in Table 3 must be justified against observed vertical microphysical profiles.

*R: Flight Patterns - From this comment, we realized the need to include a more detailed explanation about the datasets from the G1 aircraft, the creation of Fig. 3 of the manuscript, and the subsequent modeling of warm clouds from the statistics shown in Table 3 of the manuscript. Therefore, we clarify that, for both IOPs, only flights that flew over the area  $60.75^\circ W, 3.3^\circ S, 60.45^\circ W, 3.1^\circ S$  (see Figures 8 and 9 below) were considered, showing that the pattern of flight selection was consistent between the two IOPs. In addition, we also clarify that the medians shown in Figure 3 of the manuscript included experimental points from all altitudes where clouds with liquid water (zero ice content in the cloud) were identified and for which  $N_d \geq 0.3$  particles/ $cm^3$  and  $LWC \geq 0.02$  g/ $m^3$ , also seeking to avoid points related to fog. Although no filter was applied to distinguish between cumuliform and stratiform clouds, these clouds were selected using the same filters in both IOPs. Vertical Profiling - While the  $re_{ff}$  medians in Fig. 3 consider only the previously mentioned filters, the  $re_{ff}$  percentiles shown in Table 3 of the manuscript were calculated from the application of the equations shown in the legend of Fig. 3 and the subsequent application, to the CTH and LWC data measured by ground instrumentation, of the filters  $CTH \leq 3$  km and  $0.2$  g/ $m^3 \leq LWC_{ground} \leq 0.4$  g/ $m^3$ . This shows that the  $re_{ff}$  percentiles in Table 3 do not correspond to specific flight levels, but rather to a statistical representation, by layer, of cumuliform and stratiform clouds. As pointed out in the second paragraph of section 2.6 of the manuscript, there is literature*

showing that the reff of liquid clouds increases with altitude in the atmosphere (e.g., Martins et al., 2011; Rosenfeld and Woodley, 2003), justifying the use of the median to represent the middle layer of simulated clouds in LibRadtran. This discussion was included in the legend of Figure 3 and in sections 2.5 and 2.6 of the revised version of the manuscript.

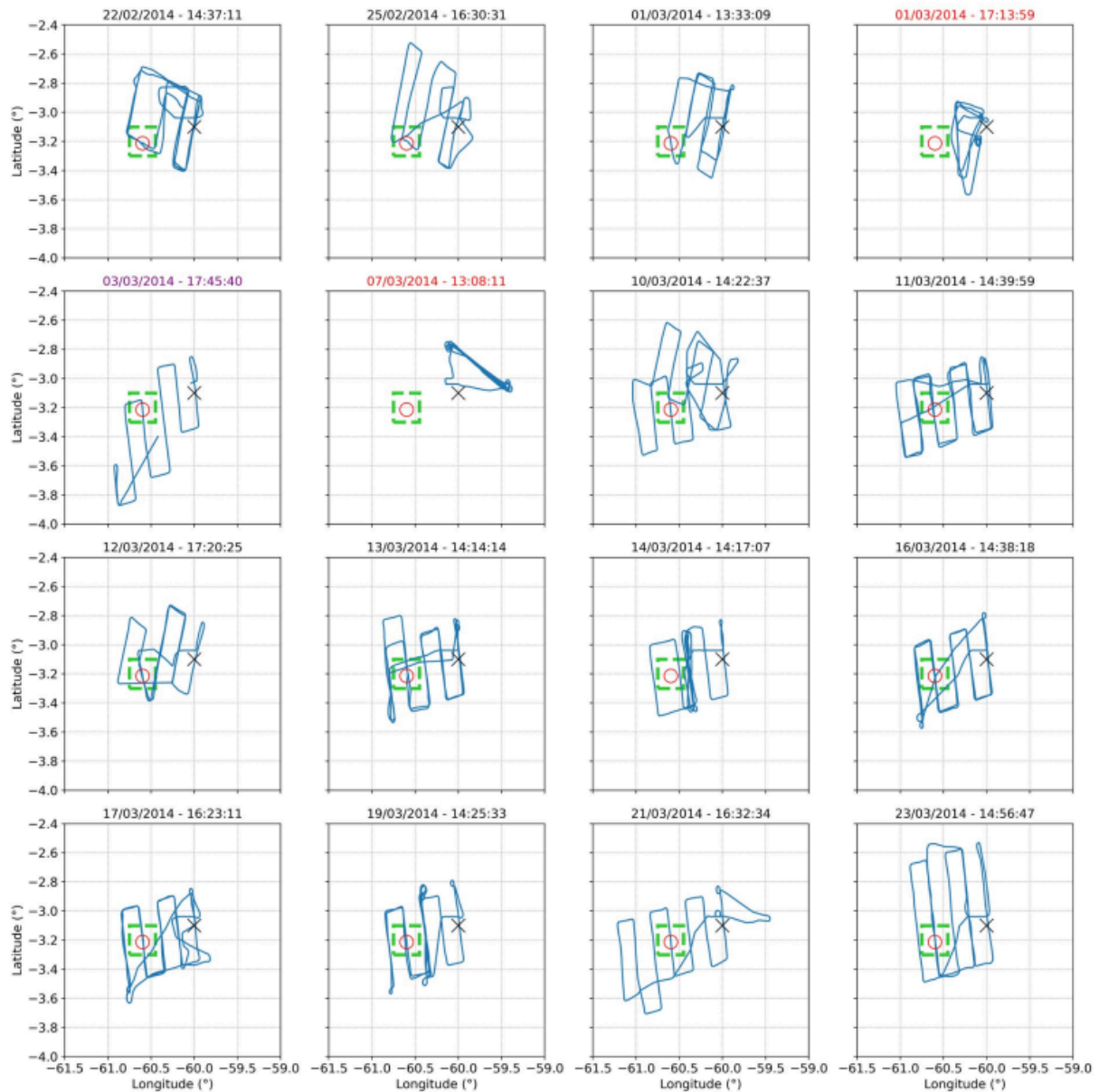
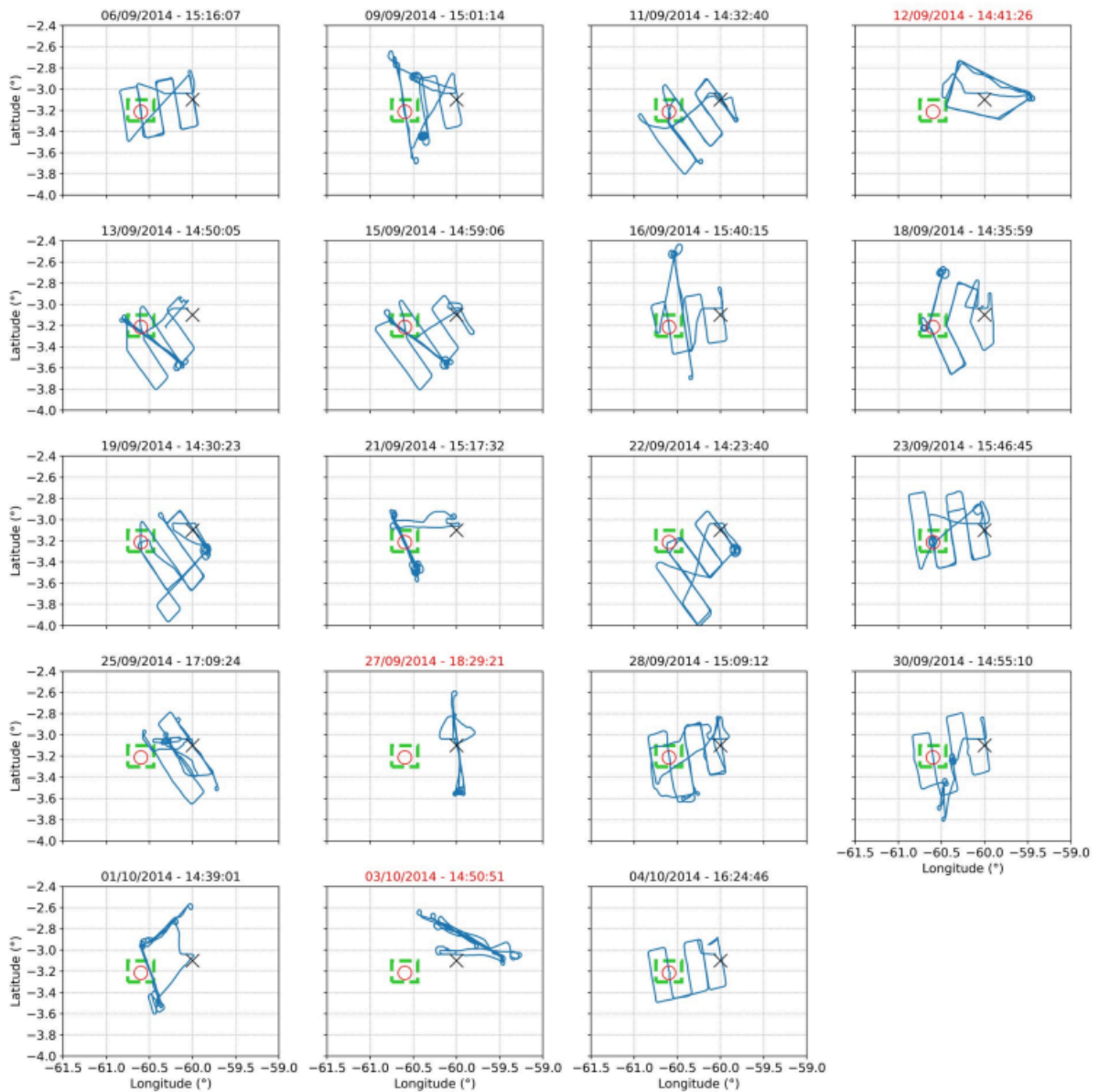


Figure 8: Daily trajectories (blue lines) of all flights performed by G-1 during IOP 1. The "X" indicates the geographic location of the G-1's starting point (Manaus), while the red circle shows the location of site T3. The area inside the dotted green line (60.75° W, 3.3° S, 60.45° W, 3.1° S) indicates the study region; that is, only flights that crossed this delimited area around T3 (plots with black titles) had their data considered.



*Figure 9: Daily trajectories (blue lines) of all flights performed by G-1 during IOP 2. Only the data relating to the plots with black titles were used.*

**4) Radiative Transfer Model Configuration:** The description of the libRadtran setup in Sections 2.6 and 2.7 is insufficient for reproducibility. Input Requirements: While LWC and reff are cited, it is unclear if the model requires the full droplet size distribution (Nd) as an input. Isolation of Aerosol Effects: The authors must explain how they isolate the aerosol induced change in optical properties from changes in other macroscopic cloud properties (e.g., cloud base/top heights) during the IRF calculation.

*R: Input Requirements - We agree that the description of the simulation setup needs to be further developed, which was done in section 2.7 of the revised version of the manuscript. Considering this, typical examples of input files (extension .inp) used in LibRadtran are available at <https://doi.org/10.5281/zenodo.20610494>. This link also includes a compiled folder containing all the datasets used in the study and was included in the "Data availability" section of the revised version of the manuscript. In all .inp files, it is evident that LibRadtran does not require the complete droplet size distribution (Nd) as input. This observation was included in the title of Table 3 in the new version of the manuscript. Isolation of Aerosol Effects - As explained in the answer to the major question 2), the use of a fixed temperature profile sought to avoid considering seasonality due to the thermodynamic variation in the macroscopic properties (LWP, CBH, CTH) of the analyzed clouds. The effects of seasonal contamination due to dynamics on these same properties are embedded in their statistical distributions. The isolation of changes in reff and LWC due to aerosol-induced changes occurs, as explained in the answer to major question 1), because the values of Nd and reff vary differently between IOPs for a fixed LWC bin. More detailed discussions on this point are included in sections 2.8 and 4.*

#### **Answers to the minor comments:**

1- Line 80 and 87: The Value-Added Products (VAPs) should be formally identified. The cloud fraction product from Riihimaki et al. (2019) is the RADFLUXANAL VAP, and the cloud boundary product is the ARSCL (Active Remote Sensing of Clouds) VAP.

*R: Done. Changes are available in the revised version of the manuscript.*

2- Line 84-85: Clarify if the Liquid Water Path (LWP) was self-derived or sourced from the MWRRET VAP. Please provide the estimated uncertainty for the LWP retrieval, as it directly impacts the LWCground calculation.

*R: Done. Changes are available in the revised version of the manuscript.*

3- Figure 1 includes CCN but omits AOD, while Figure 2 includes AOD but omits CCN. Both parameters are essential for a complete assessment of the scenario identification.

*R: We reconfigured Figure 1 to include the AOD values measured by AERONET during 2014. In Figure 2, CCN values were not considered because, for most of 2015, the measurements made by CCNPC were classified as incorrect by the ARM quality flags. We included this explanation in the legend of Figure 2.*

4- Figure 2: Address the sparse nature of the MFRSR AOD data outside the day 150-270 window.

*R: Done. A comment has been added to the caption of Figure 2.*

5- In Section 2.5 (Lines 221-225), provide more detail on how the reff database was extended.

*R: From this comment, we noticed an inaccuracy in the text. Therefore, we clarify that the reff database derived from in-situ measurements was not extended, but rather used to obtain the adjustments shown in Figure 3 (of the manuscript), from which new reff values were derived. This textual inaccuracy was corrected at the beginning of paragraphs 2 and 5 of section 2.5.*