Title: Observed Impacts of Aerosol Regimes on Energy and Carbon Fluxes in the Amazon Forest

Response (blue color) to anonymous Referee #1 (black). The original manuscript was changed accordingly. The lines indicated in our answers correspond to the track version of the manuscript.

General comment

The manuscript investigates how contrasting aerosol optical depth (AOD) regimes affect surface energy and carbon fluxes over an undisturbed Amazon rainforest using long-term in situ data (2016–2022) from the Amazon Tall Tower Observatory (ATTO). The authors focus on differences between "clean" (AOD < 0.13) and "polluted" (AOD > 0.40) regimes and assess impacts on radiation, sensible and latent heat fluxes, and CO_2 exchange. The topic is highly relevant to ACP because it addresses aerosol–biosphere–atmosphere interactions in one of the planet's key ecosystems. The study provides new observational insights from a unique long-term dataset and uses appropriate statistical tools (Spearman correlation, Pillai's trace, Random Forest) to assess nonlinear relationships. I think the paper is well written and it is neatly exposed. The literature cited is adequate.

We would like to thank Referee #1 for their detailed review of our manuscript and for the positive feedback. We are also grateful for the valuable contributions that helped to clarify the text and refine the analyses.

The manuscript presents an interesting empirical analysis of aerosol effects on energy and carbon fluxes in the Amazon. However, the methodology lacks quantitative robustness in **(i)** defining aerosol pollution regimes and in assessing statistical significance of differences between them and improved discussion. The structure and figures are generally clear, but the **(ii)** discussion often repeats background concepts and lacks a mechanistic synthesis connecting radiation, energy partitioning, and ecosystem carbon exchange.

We thank the Reviewer #1 for these important comments. They will certainly help to improve the methodology and discussion of the results.

We would like to begin our responses by stating that in the new version of the manuscript, we regrouped our data in a way that allowed us to include a greater number of runs (half-hour periods). In the previous version of the manuscript, in addition to excluding all periods when clouds were present, which is very common in the Amazon, we also excluded all data from a given day and time when a variable was missing. For example, if we did not have the reflected shortwave radiation measurement for a given time, we removed all other variables for that same time. This resulted in only 523 valid half-hour periods (370 dry season, 153 wet). In the new version of the Manuscript, we decided to regroup the variables so that they did not depend on each other. We first identified the periods in which we had the Clean and Polluted regimes (AOD < 0.13 and AOD > 0.40) and then identified how many runs of each variable were available for each regime. After this procedure, the number of runs increased substantially, as shown in Table R1, comparison between the dataset used in the first version of the manuscript (single database) and the dataset used for this new version (database by variable).

Table R1: Number of runs (half-hour periods) after all quality controls mentioned in section 2.2.

Variables	Single database					Database by variable				
	10:00 -14:00 LT		07:00 -17:00 LT		Total	10:00 -14:00 LT		07:00 -17:00 LT		Total
	No. Clean	No. Polluted	No. Clean	No. Polluted	No. Sample	No. Clean	No. Polluted	No. Clean	No. Polluted	No. Sample
SWin(Wm^{-2})	98	81	219	151	370	301	204	736	459	1195
SWout(Wm^{-2})	98	81	219	151	370	301	204	736	459	1195
LWatm(Wm^{-2})	98	81	219	151	370	301	200	733	453	1186
LWterr(Wm^{-2})	98	81	219	151	370	301	204	735	459	1194
Rn(Wm^{-2})	98	81	219	151	370	301	200	733	453	1186
H(Wm^{-2})	98	81	219	151	370	197	192	455	389	844
LE(Wm^{-2})	98	81	219	151	370	183	180	447	386	833
FCO_2($\mu molm^{-2}s^{-1}$)	98	81	219	151	370	247	195	596	405	1001
G(Wm^{-2})	98	81	219	151	370	301	218	741	487	1228

In the new version of the manuscript, we rewrote L107:

"*After filtering, the resulting dataset is summarized in Table S1 and S2*."

(i) In the previous version of the manuscript the classification into "Clean" and "Polluted" regimes was based on the 10th and 90th percentiles of the AOD distribution at ATTO. However, following the reviewer's recommendation, we performed a sensitivity analysis by applying the statistical test using three percentile thresholds (10th/90th, 15th/85th, and 20th/80th) to quantitatively assess the robustness of the regime separation (Table R2). The results show that the 10th/90th and 15th/85th percentile thresholds provide stronger physical contrasts between aerosol regimes, expressed as larger differences in median values of key variables, and are associated with statistically significant differences. Conversely, the 20th/80th threshold leads to a loss of statistical significance for several variables, indicating a dilution of the physical contrast between regimes. Based on these tests, the threshold was maintained to the 10th/90th percentiles, as this choice preserves physically meaningful differences between aerosol regimes.

Table R2. Results of the statistical test comparing all analyzed variables between clean and polluted aerosol regimes for three AOD thresholds. Panels correspond to: 10th–90th percentiles (top), 15th–85th percentiles (middle), and 20th–80th percentiles (bottom).

Main Stage - Percentil 10 (AOD≤0.13), Percentil 90 (AOD≥0.40)

Variable	N Clean	Mean Clean	SD Clean	N Polluted	Mean Polluted	SD Polluted	U Statistic	p-value	Significance
SWin(Wm^{-2})	301	836.5	165.2	204	813.5	124.4	35391	0.004	**
SWout(Wm^{-2})	301	92.8	19.7	204	95.9	15.1	27859	0.077	ns
LWatm(Wm^{-2})	301	431.5	10.4	200	432.1	9.4	29439	0.677	ns
LWterr(Wm^{-2})	301	484.7	14.0	204	483.6	10.8	34148	0.032	*
Rn(Wm^{-2})	301	659.3	137.8	200	632.8	100.8	35671	0.000	***
H(Wm^{-2})	197	160.6	67.8	192	138.9	61.4	22611	0.001	***
LE(Wm^{-2})	183	426.7	136.8	180	417.8	146.7	17246	0.438	ns
FCO_2($\mu mol m^{-2} s^{-1}$)	247	-12.5	8.0	195	-17.4	8.6	32125	0.000	***
G(Wm^{-2})	301	1.8	1.6	218	0.8	1.4	43719	0.000	***

Wide Stage - Percentil 15 (AOD≤0.14), Percentil 85 (AOD≥0.36)

Variable	N Clean	Mean Clean	SD Clean	N Polluted	Mean Polluted	SD Polluted	U Statistic	p-value	Significance
SWin(Wm^{-2})	423	829.1	166.2	272	816.6	131.5	62510	0.054	ns
SWout(Wm^{-2})	423	91.8	20.1	272	95.7	15.6	50534	0.007	**
LWatm(Wm^{-2})	423	432.7	10.0	268	433.2	9.4	55655	0.688	ns
LWterr(Wm^{-2})	423	484.1	14.5	272	484.5	11.0	59785	0.382	ns
Rn(Wm^{-2})	423	654.7	138.0	268	634.7	107.7	64197	0.003	**
H(Wm^{-2})	281	157.5	70.0	258	139.0	62.7	42045	0.001	**
LE(Wm^{-2})	265	422.3	133.9	241	419.8	145.7	32675	0.651	ns
FCO_2($\mu mol m^{-2} s^{-1}$)	354	-12.6	8.2	263	-16.7	8.5	60056	0.000	***
G(Wm^{-2})	427	1.8	1.6	298	0.9	1.4	83989	0.000	***

Alternative Stage - Percentil 20 (AOD≤0.16), Percentil 80 (AOD≥0.32)

Variable	N Clean	Mean Clean	SD Clean	N Polluted	Mean Polluted	SD Polluted	U Statistic	p-value	Significance
SWin(Wm^{-2})	547	822.9	172.0	372	828.2	132.4	103274	0.698	ns
SWout(Wm^{-2})	547	91.1	20.4	372	96.3	15.4	85935	0.000	***
LWatm(Wm^{-2})	544	433.4	9.8	366	433.6	9.5	98174	0.723	ns
LWterr(Wm^{-2})	547	483.7	14.6	372	485.2	11.0	100283	0.712	ns
Rn(Wm^{-2})	544	650.5	144.8	366	644.3	112.1	105788	0.109	ns
H(Wm^{-2})	377	155.9	71.0	350	140.8	64.5	74993	0.001	**
LE(Wm^{-2})	355	416.4	135.7	331	420.4	148.0	58402	0.893	ns
FCO_2($\mu mol m^{-2} s^{-1}$)	472	-12.7	8.3	364	-16.0	8.5	105109	0.000	***
G(Wm^{-2})	567	1.9	1.6	411	1.0	1.5	150612	0.000	***

We clarified this point in the revised manuscript as follows:

Section 2.3:

L134-137: "*Daily averages of AOD values were obtained to investigate seasonal variability. Our analysis distinguishes two contrasting atmospheric conditions at the ATTO site, defined as "Clean" and "Polluted" using AOD thresholds derived from the dry-season AOD distribution. The Clean and Polluted regimes correspond to the 10th (AOD ≤ 0.13) and 90th (AOD ≥ 0.40) percentiles, respectively. Further details on the seasonal aerosol analysis are provided in Section 3.1 and Table S3.*"

Section 3.1:

L201-207: "*As the main goal of this work is to investigate the impact of aerosols on surface turbulent fluxes, the analysis focuses on data from the dry season. In addition, during the dry season there is more aerosol data since the could interference is much* *less pronounced than during the wet season. Two aerosol regimes were defined based on percentile thresholds of the dry-season AOD distribution. Several percentile combinations were tested to assess the robustness of the regime separation. Based on this analysis, the 10th and 90th percentiles were selected to define the Clean (AOD ≤ 0.13) and Polluted (AOD ≥ 0.40) regimes, respectively, as they preserve physically meaningful differences between aerosol regimes (See table S1).*"

(ii) In the new version of the manuscript, we improved the methodology by adding a statistical analysis for the definition of Clean and Polluted regimes. We also improved the discussion of the results to avoid repeating background concepts.

In its present form, I recommend **major revisions** according to my specific comments before the manuscript can be considered for publication in *ACP*.

Specific comments

The study contributes observational evidence from a rare, pristine tropical forest site. The long-term dataset and the combination of aerosol and flux measurements are strengths. Nevertheless, the novelty is moderate, as the main conclusions - reduction of net radiation and turbulent fluxes under high AOD, accompanied by enhanced CO_2 assimilation - are qualitatively consistent with previous literature (e.g., Cirino et al. 2014; Braghiere et al. 2020; Palácios et al. 2022). **(i)** The novelty would be strengthened by including a quantitative analysis of diffuse versus direct radiation, or by exploring seasonally resolved patterns rather than aggregating all data into two AOD categories. **(ii)** Defining "Clean" (AOD < 0.13) and "Polluted" (AOD > 0.40) purely from percentiles is arbitrary. Include a sensitivity test or physical rationale for these cutoffs. **(iii)** To increase the scientific value of the study, the authors should demonstrate, through appropriate statistical testing, whether the observed reductions (≈10%) are robust across years and not driven by interannual variability.

(i) We thank the reviewer for their valuable comment. Diffuse radiation (SWd) measurements are available only for the year 2021 for our experimental site. Prior to this period, SWd was not measured at the ATTO site, and in 2022 the sensor experienced technical issues that affected data quality. Nevertheless, to address the reviewer's suggestion, we quantified the diffuse radiation fraction (Fd = SWd/SWin) for the available period (2021) and compared Fd between Clean and Polluted aerosol regimes. Our results indicate higher Fd values under Polluted regime compared to Clean regime (Figure R1). Specifically between 10:00 and 14:00 LT, the mean Fd values were 0.43 and 0.27 for Polluted and Clean regime, respectively, indicating an absolute difference of 0.16 between the two regimes ($p<0.05$). This is consistent with enhanced scattering of solar radiation associated with increased aerosol loading (Giorgi et al., 2002; Seinfeld and Pandis, 2016; Ezhova et al., 2018). Moreover, daytime CO_2 fluxes showed a non-linear dependence on Fd, with net CO_2 uptake increasing up to an Fd threshold (≈0.6) and decreasing at higher Fd values (Figure R2). This behaviour was consistent with the response of NEE (net ecosystem exchange) for diffuse radiation reported by Deng et al. (2022) for four forest sites in China and aligns with the global-scale mechanisms proposed by Mercado et al. (2009).

These results provide observational support for the proposed mechanism linking aerosol loading, radiation partitioning, and ecosystem carbon exchange.

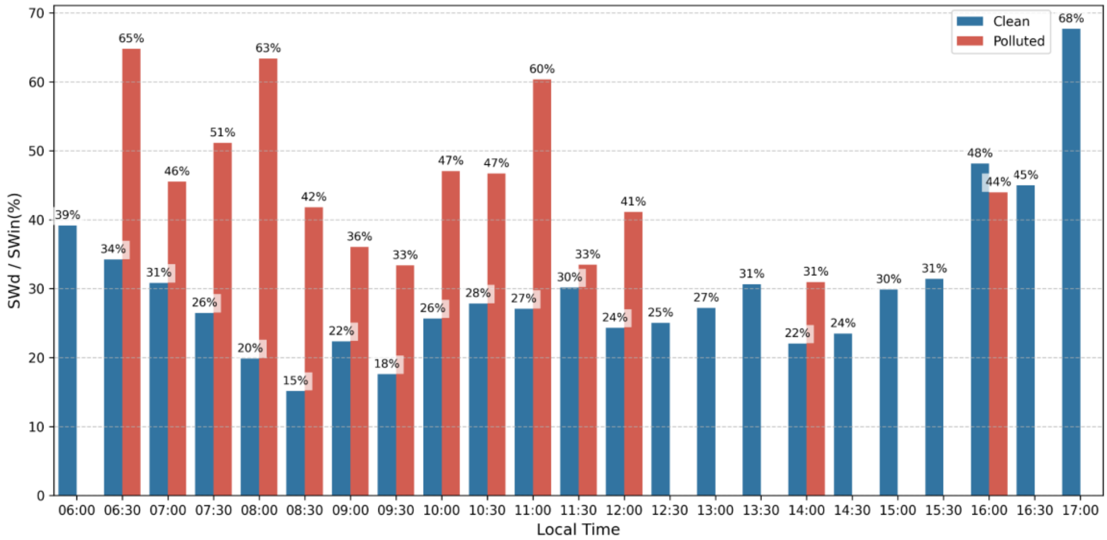

Figure R1. Diffuse radiation fraction (Fd = SWd/SWin) under Clean and Polluted aerosol regimes during 2021.

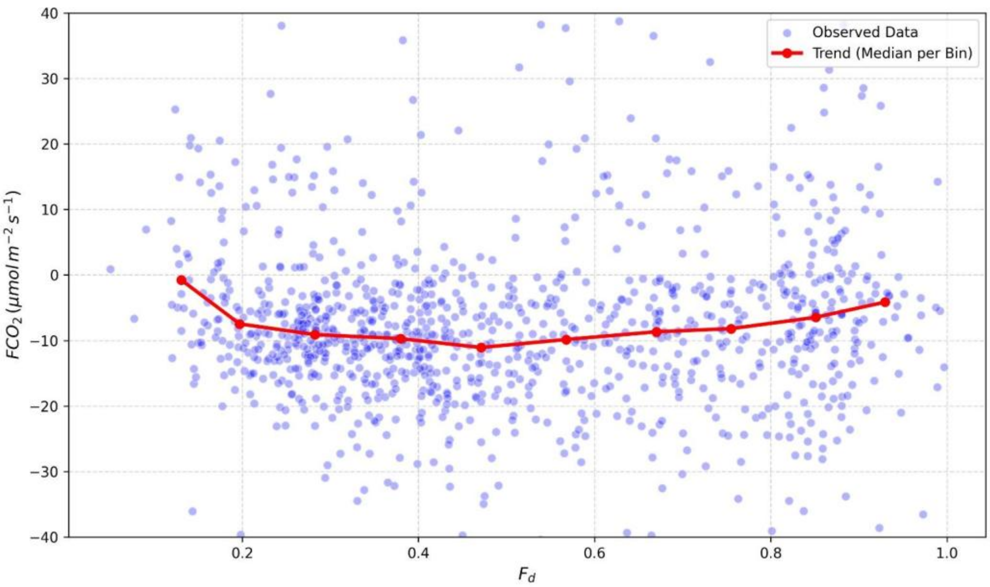

Figure R2. The relationship between CO_2 flux and diffuse radiation fraction (Fd) during 2021 at ATTO site.

The Methodology and Results sections have been updated in the new version of the manuscript as follows:

L87-90: "*Additionally, diffuse shortwave radiation (SWd) was measured using a SPN1 Pyranometer (Delta-T Devices) installed at 75 m above ground level. However, SWd data were available only for 2021, prior to this year, SWd was not measured at the ATTO site, and data from 2022 were excluded due to technical issues with the sensor.*"

L328-337: "*We quantified the diffuse radiation fraction (Fd = SWd/SWin) for the available period (2021) and compared Fd between Clean and Polluted aerosol regimes. Our results indicate higher Fd values under the Polluted regime compared to Clean regime (Figure S1). Specifically for the 10:00 and 14:00 LT interval, the mean Fd values were 0.43 and 0.27 for Polluted and Clean regime, respectively, indicating an absolute difference of 0.16 between the two regimes ($p<0.05$). This is consistent with enhanced scattering of solar radiation associated with increased aerosol loading (Giorgi et al., 2002; Seinfeld and Pandis, 2016; Ezhova et al., 2018). Moreover, daytime CO_2 fluxes showed a non-linear dependence on Fd, with net CO_2 uptake increasing up to an Fd threshold (~0.6) and decreasing at higher Fd values (Figure S2). This behaviour was consistent with the response of net ecosystem exchange for diffuse radiation reported by Deng et al. (2022) for four forest sites in China, and aligns with the global-scale mechanisms proposed by Mercado et al. (2009). These results provide observational support for the proposed mechanism linking aerosol loading, radiation partitioning, and ecosystem carbon exchange.*"

(ii) We assessed the robustness of our results in relation to different regime thresholds as detailed in the General comment (L59-71 and Table R2 of this document).

(iii) We analyzed 2020 and 2022 separately due to higher data availability, while the remaining years (2016, 2017, 2018, 2019, and 2021), which had lower data availability, were grouped into a single category (Remaining). The pattern observed in the preliminary version of the manuscript (Figs. 5 and 6) was consistent across individual years (Figure R3). That is, under polluted conditions, reductions in SWin lead to a decrease in Rn, followed by decreases in H, LE, G, and FCO_2.

The following sentence has been added to the revised manuscript.

L293: "The reductions in H, LE, G, and FCO_2 shown in Fig. 6 and Fig. 7 were also observed across individual years (see Fig. S3)."

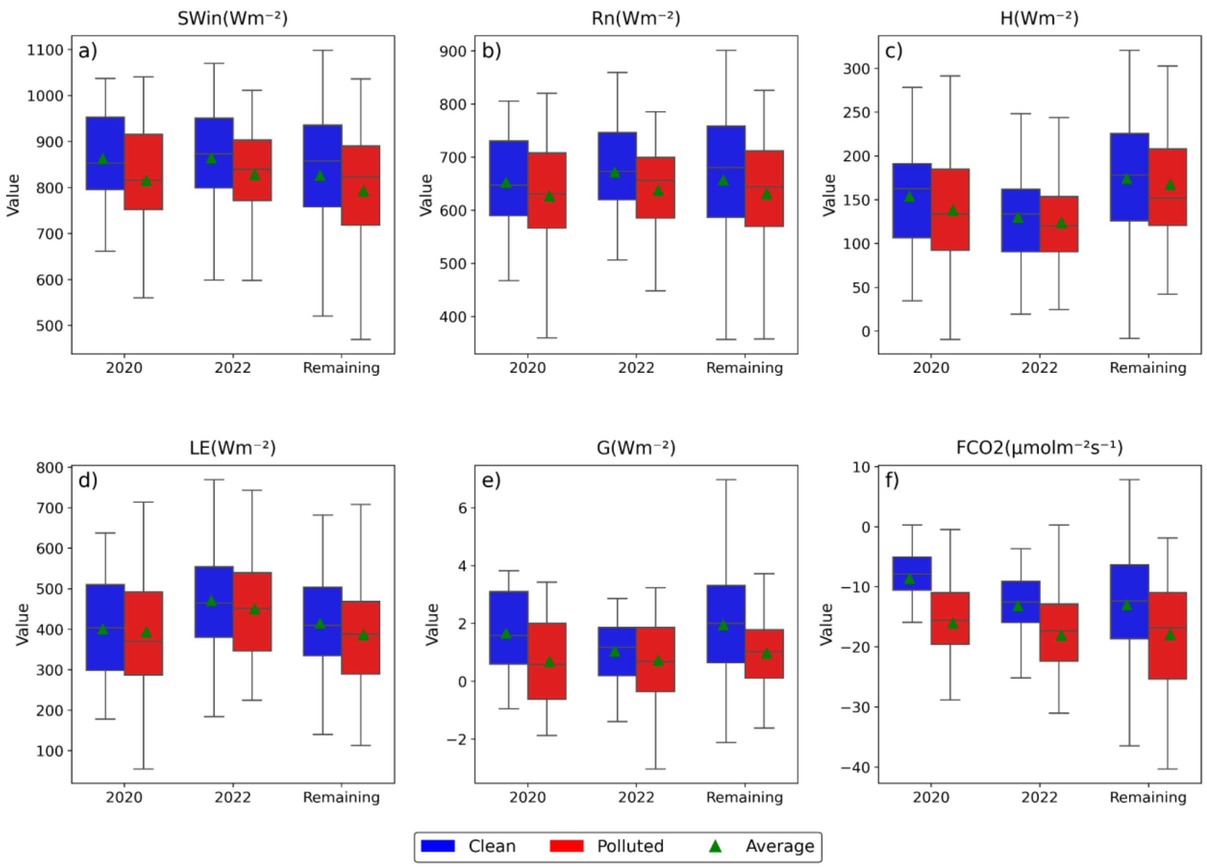

Figure R3. Box plots of a) incoming shortwave radiation (SWin), b) net radiation (Rn), c) sensible heat flux (H), d) latent heat flux (LE), e) ground heat flux (G), f) CO_2 flux (FCO_2). All variables under clean (blue) and polluted (red) aerosol regimes for 2020, 2022, and the remaining years (2016, 2017, 2018, 2019 and 2021), grouped due to limited data availability. Triangles indicate the mean value.

Line 94-97. Only 523 valid half-hour periods (370 dry season, 153 wet) are quite small relative to the six-year period. The statistical representativeness and interannual variability need further discussion.

We modified our methodology to obtain as much data as possible, as detailed in lines L36-50 and Table R1 of this document.

Line 173-174. The section on radiative fluxes should include a graph of the full diurnal cycle of SW, LW, and Rn to visually demonstrate the 10:00 - 14:00 LT maximum. This would strengthen the rationale for focusing on that time window.

We thank Reviewer #1 for this valuable comment. As recommended, we have included Figure R4 in Section 3.2 of the manuscript (Figure 4 in the new version of the manuscript), which shows the full diurnal cycles of shortwave (SW), longwave (LW), and net radiation (Rn), and highlights the maximum between 10:00 and 14:00 LT.

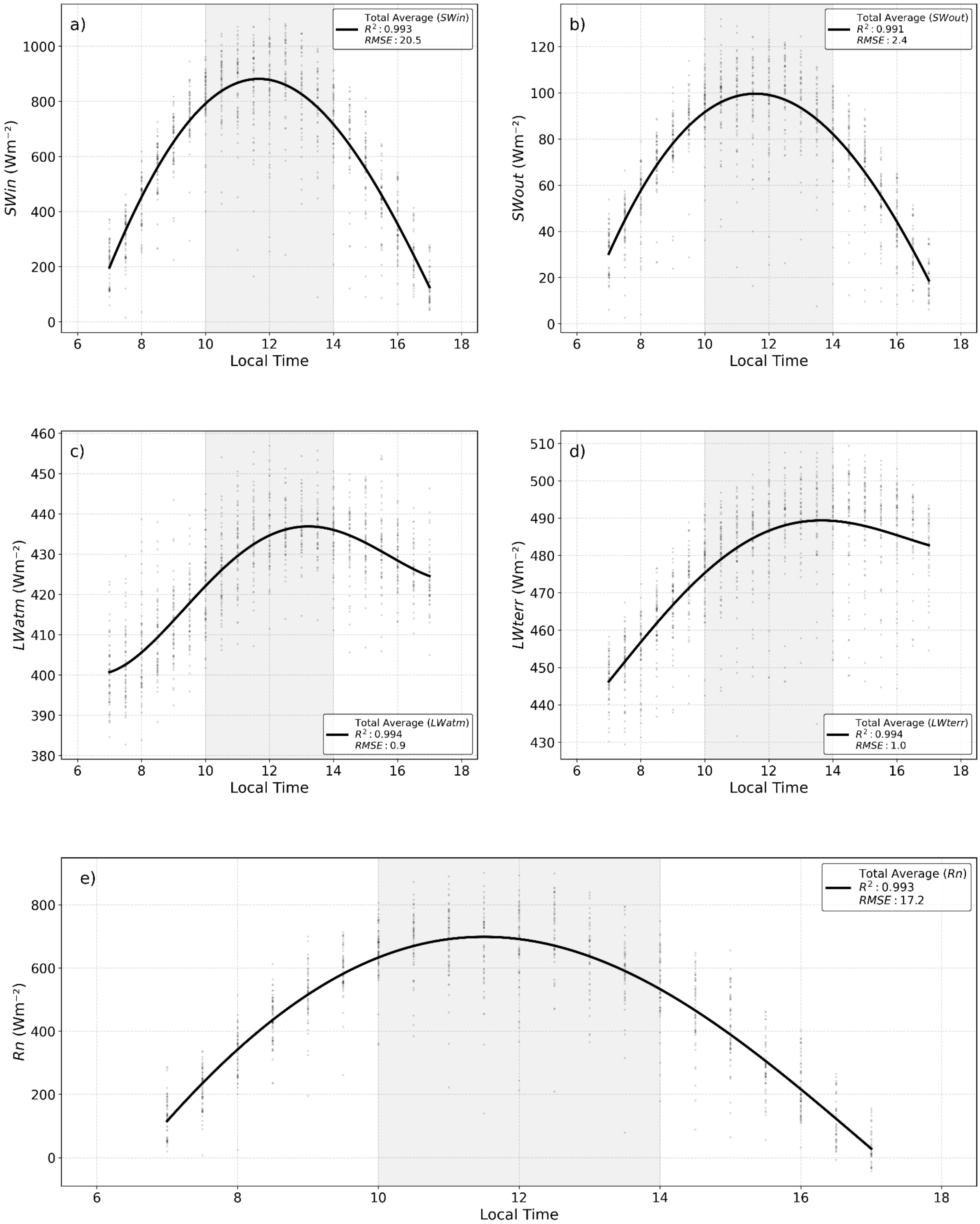

Figura R4. Diurnal cycles of radiative fluxes during the dry season from 2016 to 2022: (a) incoming (SWin) and (b) outgoing (SWout) shortwave radiation, (c) incoming atmospheric (LWatm) and (d) outgoing terrestrial (LWterr) longwave radiation, and (e) net radiation (Rn). Markers indicate observed data, and solid lines represent fourth-order polynomial fits, with the corresponding R^2 and RMSE.

In the revised version of the manuscript, we have added Figure R4 and the following paragraph to clarify the choice of the analysis period:

L209-212 : *"As described in Section 2.3, the comparisons between Clean and Polluted regimes were restricted to the 10:00–14:00 LT period, corresponding to the maximum net radiation. The full diurnal cycles of shortwave, longwave, and net radiation during the dry season (2016–2022) show that the maximum values occur between 10:00 and 14:00 LT (Figure 4), supporting the choice of this time window for the subsequent analyses*."

Line 203-204. The physical interpretation of the longwave radiation components (LWatm and LWterr) is interesting, but it would benefit from quantitative support - for instance, by including a vertical temperature profile or an estimate of surface and atmospheric emissivity.

We used the Stefan–Boltzmann equation: LW = $\varepsilon\sigma T^4$, where LW is the longwave radiation (Wm^{-2}), ε is the emissivity, σ is the Stefan–Boltzmann constant (5.67×10^{-8} $Wm^{-2}K^{-4}$) e T is the absolute temperature (K).

Using this relationship, separately for the Clean and Polluted regimes, we estimated the surface emissivity (εs) from the measured outgoing longwave radiation (LWterr) and the infrared surface temperature (Ts), and the atmospheric emissivity (εa) from the measured incoming longwave radiation (LWatm) and air temperature (Ta). The resulting emissivity values are shown in Table 3. The emissivities exhibit very similar values under both regimes, suggesting that the differences in aerosol conditions were not sufficient to affect the surface emissivity or the atmospheric emissivity.

Table R3: Mean values of incoming and outgoing longwave radiation (LWatm and LWterr), air temperature (Ta), surface infrared temperature (Ts), and the corresponding atmospheric (εa) and surface (εs) emissivities, under Clean and Polluted regimes for the 10:00 and 14:00 LT interval.

	LWatm (Wm^{-2})	LWterr (Wm^{-2})	Ta (°C)	Ts (°C)	εa	εs
Clean	431.5	484.74	30.3	32.6	0.898	0.978
Polluted	432.1	483.60	30.0	31.7	0.902	0.988

Line 227-232. The manuscript would benefit from a discussion of the energy balance closure, specifically addressing the discrepancy between Rn and the sum of H, LE, and G. Reporting the residuals for both clean and polluted regimes would provide a clearer evaluation of data quality and potential systematic biases.

We thank the reviewer for this important comment. Following this suggestion, we updated the manuscript by adding the following text:

L278-281: "*The surface energy balance closure was 0.89 for the Clean regime and 0.88 for the Polluted regime, comparable to values reported in the literature (Mauder et al., 2024). The corresponding residuals were of similar magnitude (70 Wm^{-2} for Clean and 75 Wm^{-2} for Polluted), indicating that the observed differences in energy fluxes are not related to differences in energy balance closure.*"

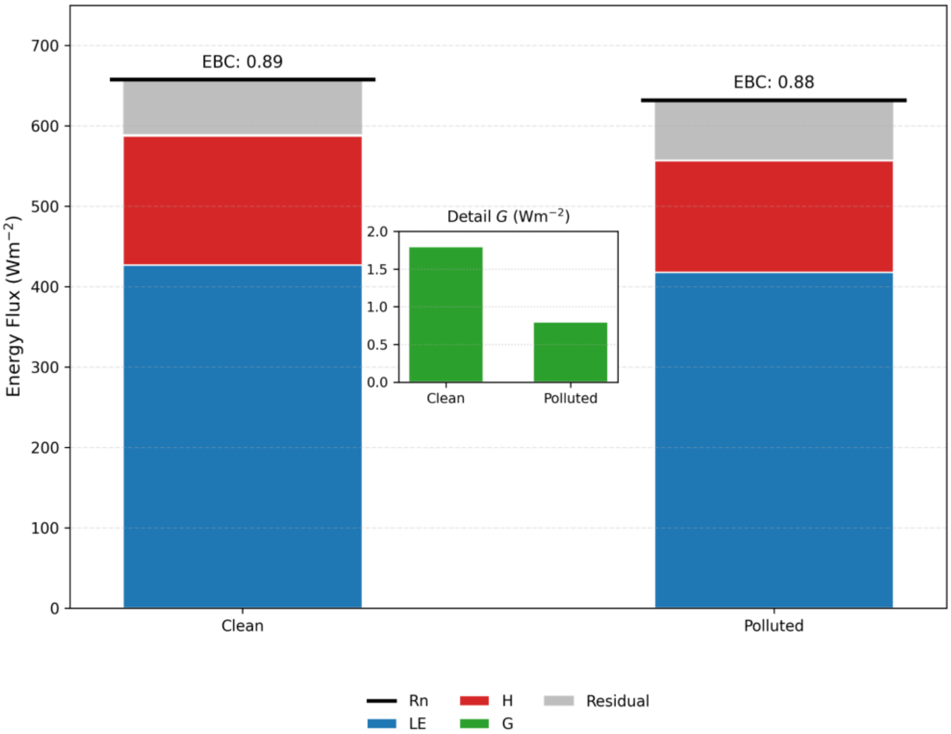

Figura R5. Energy balance closure for Clean and Polluted regimes during the dry season from 2016 to 2022, considering the 10:00–14:00 local time.

Figure 6. The fourth-order polynomial fits to the diurnal cycles provide a useful visual comparison, but the authors should complement them with statistical analyses to confirm that the apparent differences between regimes are statistically significant.

We thank the reviewer for this comment. The statistical analyses comparing Clean and Polluted aerosol regimes were performed for all analyzed variables, including those shown in Figure 6. The results are presented in Table R1, as requested in the General Comments.

Line 250-255. The connection between aerosol effects and water-use efficiency (WUE) is largely speculative because WUE is not quantitatively evaluated in the manuscript. The authors should consider calculating WUE (for example, as GPP/ET using FCO_2 and LE data) or presenting an appropriate proxy to substantiate this aspect of the discussion.

We thank the reviewer for this important comment. Following this suggestion, we now quantify water-use efficiency (WUE) using $|FCO_2|$ / LE as a proxy, and performed statistical analyses comparing Clean and Polluted aerosol regimes (Table R4).

Table R4: Mean water-use efficiency (WUE) calculated as $|FCO_2|$ / LE (μmolJ^{-1}), under polluted and clean regimes for individual years (2020 and 2022), the remaining years (2016, 2017, 2018, 2019 and 2021) and all years combined.

Year	AOD Regime	$\lvert FCO_2 \rvert$ / LE ($\mu molJ^{-1}$)
2020	Clean	0.022
2020	Polluted	0.042
2022	Clean	0.031
2022	Polluted	0.044
Remaining	Clean	0.026
Remaining	Polluted	0.049
All Years	Clean	0.029
All Years	Polluted	0.042

In the new version of the Manuscript we added the follow sentence:

L304-307: "*In this study, WUE was estimated using FCO_2/LE as a proxy. WUE was significantly higher under Polluted compared to Clean regime(mean values of 0.042 and 0.029, respectively, $p < 0.05$). This indicates that under Polluted regime, vegetation assimilates more carbon per unit of water lost, consistent with the observed reduction in latent heat flux (Figure 6) despite enhanced CO_2 uptake (Figure 7).*"

Line 242-245. It seems to me that there is some inconsistency throughout the manuscript regarding the sign convention of CO_2 flux. The authors should clearly state that CO_2 uptake by the ecosystem corresponds to a negative flux, while positive flux values indicate a CO_2 emission to the atmosphere. Accordingly, a "drop" or decrease in FCO_2 should represent reduced carbon uptake, not enhanced assimilation. In the Abstract, for example, Authors should clarify the meaning of "decrease in CO_2 fluxes by 58%" (does this mean more negative flux, i.e., greater uptake?). Clarifying this point is essential for avoiding misinterpretation of the results and ensuring consistency across figures, tables, and the discussion.

We thank the reviewer for pointing out this important issue. To avoid any ambiguity, we have clarified throughout the manuscript that negative CO_2 fluxes indicate net ecosystem uptake. The text has been revised as follows:

L290-292: "*In addition to their effect on energy fluxes, aerosols were found to have a significant influence on the CO_2 flux, becoming more negative by an average of 4.9 $\mu mol\ m^{-2} s^{-1}$ (39.5 %) in the polluted regime compared to clean conditions between 10:00 and 14:00 LT.*"

Abstract: "*We find that enhanced aerosol presence reduces both sensible heat flux and energy available for evapotranspiration by approximately 13.5 % and 2.1% respectively, while increasing CO_2 uptake (i.e., CO_2 flux becoming more negative) by about 39.5 %.*"

The figures are generally clear and well designed, but they would benefit from the inclusion of confidence intervals or error bars to convey the statistical variability of the data. Adding uncertainty information would allow readers to better assess the robustness of the observed differences between regimes and the reliability of the fitted curves.

We thank the reviewer for this suggestion. Figures 4, 5, and 6 (Fig. 5, 6, 7 in the new version of the manuscript) have been revised to include the observed data points underlying the averaged curves, allowing a direct visualization of the data variability.

Minor comments

All physical variables (Rn, H, LE, FCO_2, AOD, etc.) should be written in italics or formatted with the equation editor for consistency and readability.

The entire manuscript was revised to ensure consistent formatting of all physical variables. Thanks.

Throughout the manuscript, several acronyms are not explicitly defined (ARF24h, LWterr), which may affect readability. I recommend defining each acronym upon first use.

The entire manuscript was revised to ensure that all acronyms are explicitly defined at their first occurrence. Thanks.

Line 191. “ARF24h”, did Authors refer to daily mean? It should be clarified

Yes, in the revised manuscript, we have removed the term *ARF24h* to avoid ambiguity as follows:

L227-230: “*Consistent with these findings, Palácios et al. (2022) estimated an average ARF of −20.77 ± 5.04 Wm^{-2} for the dry season in the central Amazon. Procopio et al. (2004) found daily ARF values ranging from −21 to −74 Wm^{-2} in the deforestation arc, an area with higher levels of pollution than the central Amazon. Rizzo et al. (2011) investigated this central region and reported a daily ARF value of −32 Wm^{-2}.*”

Line 187-188. The phrase “In contrast” seems used incorrectly; the studies cited do not contradict one another, showing similar ARF values (within the estimated errors). The Authors should revise wording.

We thank the reviewer for this comment. The term “*In contrast*” was replaced by *“Consistent with these findings*” as detailed in previous comment (L352-356 in this document).

Table3. Caption. FCO should be replaced by FCO2

The caption of Table 3 has been revised.

Line 299. As before, CO -> CO2

The text has been updated accordingly.

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