REVIEWER COMMENTS TO AUTHOR

The authors thank the reviewers for their useful comments. We recognize that some manuscript sections were too long and repetitive. We improved the manuscript by reducing and synthesizing all the sections. In addition, we added a co-author (C. Mouchel-Vallon) to this paper who helped significantly in this review process, especially in the acid contribution section, and in the global rewriting of the paper.
Answers and explanations are given here, accompanied by detailed descriptions of the modifications brought to the manuscript. As suggested by Reviewer #1, we moved some content to newly created appendix and supplementary material.
Additionally, we revised the abstract and the conclusion to better reflect the content of the paper.

Reviewer#1

General comments

General comment 1: The paper is too long and very descriptive, sometimes repeating the details several times. This made the manuscript hard to read. The results section should be shortened, and the conclusions should be more explicit. I would suggest reducing the total manuscript's content by at least 40-50%. Please, move some information and details to supplementary material.

Response to general comment 1:
We have reduced drastically the manuscript. We rewrote some parts of text; we sent some parts of text and figures and tables in supplementary materials and we deleted a section.
We deleted the section Monthly and Seasonal Variations (195 lines) now integrated in the results sections presenting all the contributions.

Globally, the paper previously represented 826 lines against 675 lines in this reviewed version. The Material and Method section has been reduced from 301 lines to 192 lines. The results section has been reduced 408 lines to 366.

General comment 2: The authors could explore the application of statistical dimension reduction techniques, such as partial least squares or principal component analysis, which could help to improve the results by reducing individual analysis and the conclusions of the study.
Response to general comment 2:
We have performed the requested PCA analysis. Results are presented at the end of this document. As major findings don't constitute an added value compared to our calculations of correlation coefficient, we decided not to include this ACP analysis in the text.

General comment 3*:* The quality of the figures is not good enough. The authors should improve them by increasing the quality resolution and the size of the text used. Additionally, the captions need to be more detailed.

Response to general comment 3: We have improved quality resolution and the size of the text used in figures and try to detail more specifically captions.

General comment 4: References need to be incorporated in the discussion of the results.
Response to comment 4:
We have added references in the discussion part and this can be seen in revised manuscript:
Line 138
Line 518

General comment 5: A discussion about the limitations of the study is missing. Even if there is a lack of data in the continent, and some points are mentioned at the end of the conclusions, some discussion about the limitations and the improvements for future studies should be addressed.
Response to general comment 5 We have addressed this issue in the conclusion.

General comment 6: The abstract and conclusions need to be improved. They are repetitive and seem more like a summary than proper abstract/conclusions
Response to general comment 6: We have improved abstract and conclusions.

Minor comments reviewer#1

Line 27: VWM, please add first the definition of the abbreviation
Now line 30: VWM has been defined, we write Volume Weighted Mean (VWM)

Figure 1: The resolution and text size are not good enough. Please improve it.
We modified Figure 1 to improve the quality

Lines 68-72: Too long. Please split the sentence.
We split the sentence as followed: *"In this context, the most recent study is the global assessment of precipitation chemistry and deposition carried out under the auspices of the World Meteorological Organization (WMO) - Global Atmospheric Watch (GAW) program. It aims to characterize precipitation chemical composition and to quantify deposition fluxes (wet, dry, total) of sulfur, nitrogen, acidity, sea salt, organic acids and phosphorus at global and continental scales."* Now line 73-77

Line 73: delete – before 2005 Done

Lines 94-99: Too long. Please split the sentence.
We modified the sentence to split and shorten as followed: *"In the context of the rapid urbanization and demographic explosion in Africa, it is important to improve the understanding of urban atmospheric composition and the potential impacts of air pollution in developing countries' megacities (United Nations, Department of Economic and Social Affairs, Population Division 2017; Kaba et al. 2020)".* Now line 97-100

Lines 107-111: Too long. Please split the sentence.
The sentence has been shortened considering that all information for the programs are included in Gnamien et al, 2021 and in the INDAAF web site. "*This work was carried out within the framework of the Air Pollution and Health in Urban Areas program (PASMU) implemented in 2018 (Gnamien et al., 2021); and the INDAAF program*". Now line 108 -110

Lines 112-114: Redundant, already said in lines 105-107
The sentence has been removed and some elements complete now the sentence in lines 105-107.

Line 138: delete (before Fall Done

Lines 137-143: Hard to read; please reformulate

Following the comment of reviewer 2, we decided to remove the paragraph that defined connectors. In the aim to globally shorten the paper, we decided that the information given in lines 137-143 was not absolutely necessary.

Line 174: and elsewhere, Intertropical convergence zone (ITCZ)
OK, Line 174 (now line 258) we wrote Intertropical convergence zone (ITCZ) and then we just use the abbreviation ITCZ

Line 178: avoid repetition, the Northern, the Central and Coastal climatic zones Ok done, now line 262

Figure 2 and Lines 221 -271: This section describes observational data and part of the results. Please move to the results section or supplementary information. Also, quite repetitive about the climatic zones. That was already said in the previous paragraph. Please refer to each panel when describing Figure 2.
We decided to modify section 2: Material and Methods. Especially we modified section 2.2 now intitled Meteorological parameters (now 164). This section now only presents database and references for the acquisition of meteorological parameters.

We move the description of general climatology to the result section (section 3) and we add section 3.1 intitled "Climatology of sites" to present results of the Figure 2 (see now lines 256 - 330). This paragraph has been entirely modified.

Lines 239-240. Observations in Abidjan show a weak fluctuation of air temperature and relative humidity during the studied period (2018-2020). But the authors are only showing the data from 2019 in Figure 2. Please correct or explain.
We modified the sentence to explain that meteorological observations presented in Figure a,b,c for the three sites are a mean for the period 2018-2019 (see 305-309) *"Observations in Abidjan show a weak fluctuation of mean air temperature and relative humidity over the period 2018-2020".*

Section 2.3: is quite long. Please reduce.
We tried to shorten the section, but the definition and calculation of WMO/GAW criteria are important to explain the representativity of the study's sampling and finally of our database.

Section 2: Please define organic ions.
We write now line 218-219 *"Major inorganic (Na^+, K^+, Mg^{2+}, Ca^{2+}, Cl^-, NO_3^- SO_4^{2-}, NH_4^+) and organic, derived from carboxylic acids, ($HCOO^-$, CH_3COO^-, C_2H_5 COO^-, $C_2O_4^{2-}$) ions were determined by Ionic Chromatography…"*

Line 354: NO2 Done

Line 366. Please define VCDs Done "*Vertical Column Densities (VCDs)"*. Now line 237

Line 387: Crustal element of continental origin?
We change in supplementary material in section 4 by "*Ca^{2+} is selected as a reference element from crustal origin"*

Line 391, please define NSS before the equation.

We define SSF and NSSF now in supplementary material in (See S4) (previously line 390): "*Sea Salt Fraction (SSF) and Non-Sea Salt Fraction (NSSF)*".

Lines 393 and 401: The authors use potential acidity and acidic potential… are those the same? This part is not clear; please organize it better and leave some spaces between equations and text.
We re-organized this part now and we decided to place this part in the supplementary material (See S4).

Figures 3 and 4 present a very low-resolution quality. Please improve them: Figure 3 is now in supplementary material noted Figure S1, Done

Line 475 and elsewhere. Terrigenous origin> Do the authors mean mineral dust /crustal origins?
yes, Terrigenous origin we refer to mineral dust/crustal origin for this line but we have mentioned in legend that Terrigenous in urban context refers to a mixture of anthropogenic sources and crustal source.

Figure 4. The authors are comparing sources with chemical contributions in this figure. This is confusing. I do not think it is appropriate to use this classification, since one can have organic species from terrigenous or marine origin. The same is true for acidity. Also, it is not clear which ions are represented in each category.
This classification has been used in several studies (Laouali et al., 2012, 2021) ,(Bakayoko et al., 2021; Akpo et al., 2015). Moreover, we have specified in the paper that we are not comparing sources and contributions. We are trying evaluate the contribution of each group of ionic species that can come from the same source and to do this we have specified marine and non-marine fractions of the ionic species that enter in the calculation of contributions. Thus, terrigenous contribution is made up exclusively of non-marine fraction of ionic species that are recognized as having crustal origins. But as we refer to the non-marine fraction it includes the crustal and anthropogenic fraction that is why we have specified in the legend that the terrigenous contribution is a mixture of crustal and anthropogenic fraction. We will put the contributions and the different species for each contribution in the legend to make it clearer.

Line 495 and figure 5: Why are the authors using an extended period for back trajectories when the sampling period is only during 2019-2020?
The sampling period on the three sites are 2018-2020 (Table 1). It is the reason why we present air masses back trajectories according to the period 2018-2020. We just remind the reviewer that 2018 database is not used for annual precipitation characterization (because of a non-satisfactory sampling representativity) (Table 1)) but monthly 2018 VWM concentrations are calculated (according good quality indicators of quarterly PCL%, see table 1). To be clear: we only remove the first 2018 trimester (Jan-Feb-March 2018) for Abidjan and Korhogo but we calculate monthly VWM for the others 2018 months.

Lines 547-551: some references are missing for this assertion: Done (now line 518)
Ref Laouali et al., 2012

Lines 572-579: Is the Abidjan site located upwind or downwind of the urban area? Since the authors have shown a high contribution from marine air masses, could this sulphate be related to other transport other than road-transport ones (i.e. shipping emissions)?

This sulphate could effectively be originated from shipping emissions since in the calculations of the marine contribution in Abidjan, we have 7% of marine fraction. This marine part of the sulphate could come from the shipping emissions but we have no reliable method to confirm it.

Table 4. Please increase the space between the lines. Hard to read. Done
Table 4 became Table 2

Line 605: are highly correlated with r value of (r=0.79), (r=0.70), (r=0.73). All the r are redundant. Done (now line 482)

Lines 671-675 and elsewhere: when comparing the values with literature, the authors should include references, even if they are detailed in table 4. Done

Line 741: Which is the Benin site? Done (now line 604)
We precise Djougou, in Benin

Figure 7: Please correct the square brackets Done (now line 626)
Now Figure 7 is Figure 6

Figure 8: quality needs to be improved. Please explain the X in kgX.ha-1. yr-1 (also present in other parts of the manuscript). What is the t.carb variable? Done
Figure 8 is moved to the supplementary materials, it became Figure S2. tcarb is now defined in Figure S2 as followed: the total carbonates species, calculated from this equation tcarb = $10^{(pH-5,505)}$ (Kulshrestha et al., 2003).

Lines 795-797: should be moved to the section when describing figure 4. done
We consider that this part should remain in the section that deals with the acidity of rain because we address each contribution section by section so we talk about organic acids in the section dedicated to the acid contribution. That's why we consider it will be appropriate to keep this part in the acid contribution.

Lines 803-805: To evaluate organic content, the authors must address the OC or DOC content. Evaluating four or five organic ions is not enough to reach any conclusion.
In this study we specifically study organic acids, not total or dissolved organic content. Of the organic acids measured in this work, formic acid, acetic acid and oxalic acid have been identified as the most common ones in both cloud and rain waters (Sun et al., 2016; Niu et al., 2018). We therefore consider, like in previous papers (e.g. Bakayoko et al., 2021, Akpo et al., 2015), that the measured organic acids are representative of organics contribution to rain water acidity.

Lines 826-830: Could the differences be related to the limited number of ions evaluated?
We don't think that the differences could be related to the number of ions that are analyzed. The number of ions evaluated in this work is not limited, as the ions measured in this work are the major ions as shown by many other authors cited in the paper. For instance, the study of Vet et al, 2014, that represents an overview paper of reference from the GAW/WMO program, presents a global assessment of rain composition and wet deposition at the global scale considering the same major ions analyzed in our study. It may still be possible, but highly unlikely, that some of the differences could come from some unacknowledged ions.

Section 3.2 is too long and repetitive. I would suggest reducing it by at least half and moving to the first part of the results in order to avoid redundancies. Done

Table A1 and Figure A2 (should be A1) must be improved. The text is very small, and the resolution is very low. Done

There are several typos, caps lock in the middle of sentences, double spaces, etc. English need to be revised and improved.
We agree and tried to carefully review the text to correct all the typos. We also revised the English.

Table 2 has been removed from the main text and placed in Appendix (referenced Table A2).

PCA analysis results We performed a principal component analysis (PCA) over the variables describing rain composition for each studied location. The results are displayed on Fig. 1 and analyzed below.

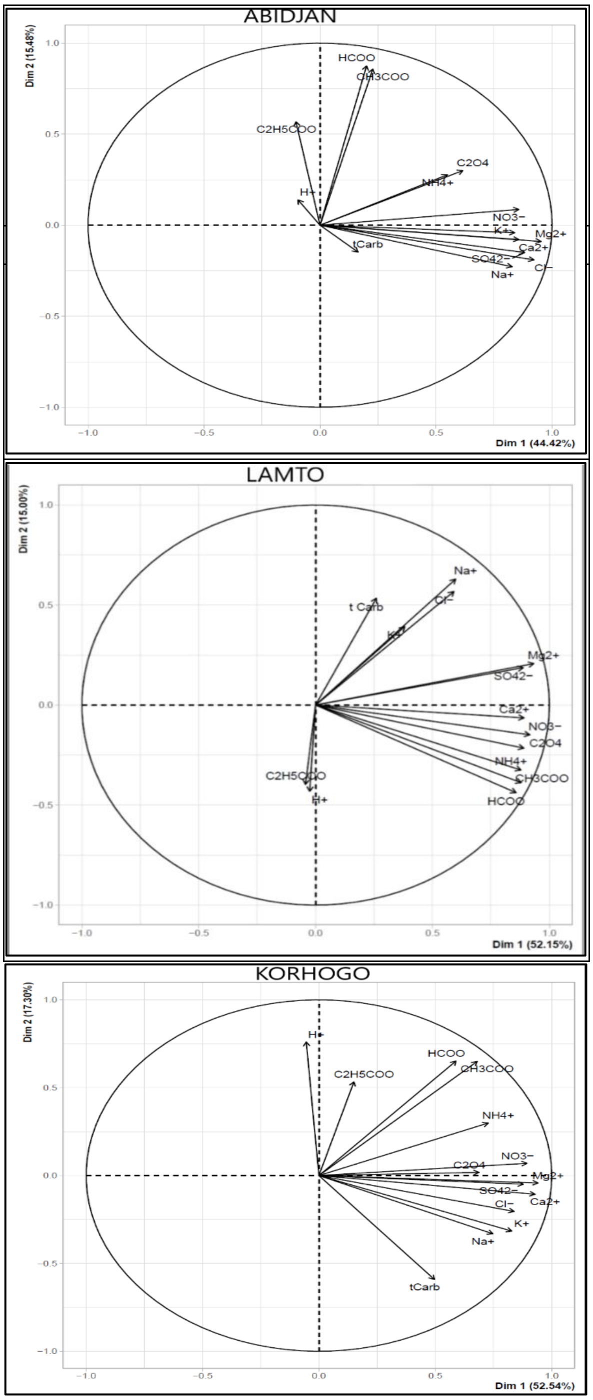

Figure 1: PCA results (*Abidjan-Lamto-Korhogo)*

In Abidjan, we note that the main factor of the PCA is composed of the variables NO_3-, K^+, Mg^{2+}, Ca^{2+}, Cl^-, SO_4^{2-}, Na^+, $C_2O_4^{2-}$, NH_4^+ which are positively grouped around the axis representing the main dimension 1 with 44.42 % of the explanatory information of the existing relations between the variables. This strong proximity between these different variables in Abidjan could be explained by the fact that these ions coming from different sources participate in chemical reaction processes that are intimately linked such as acidification and neutralization. If we couple these results with the correlation factors, we see that the variables composing the first factor all have relatively good correlations with correlations distinguishing sources such as the marine source Na^+ and Cl^- (0.94), the terrigenous source Mg^{2+} and Ca^{2+} (0.81), the anthropogenic source SO_4^{2-} and NO_3^- (0.74) and a possible fourth source (biomass fires or charcoal) with the strong correlations between K^+ and Cl^- (0.75)

The second factor of the principal component analysis in Abidjan is mainly composed of the $HCOO^-$, CH_3COO^- and $C_2H_5COO^-$ ions, with 15.48% of the explanatory information of the existing relationships between the variables. When we couple these results with the correlation factors, we see that the $HCOO^-$ and CH_3COO^- ions have a very good correlation (0.75) while the $C_2H_5COO^-$ ion shows no good relationship with the other two ions. This pattern could mean a common origin of $HCOO^-$ and CH_3COO^- ions which are the most abundant low molecular weight organic carboxylic acids in the global troposphere. They can either be emitted from direct sources such as vehicle exhaust emissions, biomass burning, biofuels, fossil fuels, and vegetation, or they can be formed in the atmosphere through photochemical reactions (Cruz et al., 2018). In the urban context of Abidjan, the most likely source could be vehicle emissions.

Lamto shows a rather different principal component analysis with the main factor of the PCA composed of the variables grouped and positively correlated on the axis representing dimension 1 with 52.15 % of the explanatory information of the existing relationships between the variables which are the following ions: Ca^{2+}, NO_3^-, $C_2O_4^{2-}$, NH_4^+, CH_3COO^-, SO_4^{2-} , $HCOO^-$. This strong correlation between these ions coming from different sources could be explained by the same mechanism that prevails at the Abidjan site, i.e. the interactions related to the acidification and neutralization process in rain water. Indeed, the very good correlation factors between the different ions (Ca^{2+} , NH_4^+) ; (Ca^{2+} , SO_4^{2-}) ; (Ca^{2+} , $HCOO^-$) ; (Ca^{2+} , $HCOO^-$) ; (Ca^{2+},Mg^{2+}) ; (Mg^{2+} , SO_4^{2-}) ; (Mg^{2+} , Cl^-) respectively 0.87, 0.76, 0.82, 0.86, 0.92, 0.88, 0.71 are an indicator of the neutralization capacity of cations on acidic compounds and according to (Lu et al. , 2011) are probably the result of the reaction of alkaline species rich in Ca^{2+} and Mg^{2+} with sulfuric, nitric, hydrochloric and organic acids.

The second factor (15.00%) is composed of Na^+, Cl^- and K^+ ions which are also contributing to dimension 1. Indeed, the strong correlation factors between Na^+ and Cl^- (0.82) and K^+ and Cl^- (0.71) confirm that these ions may come from the marine and biomass burning sources respectively (Lara et al., 2001).

Korhogo exhibits a principal component analysis on rainfall composition that is almost similar to that of Abidjan. The main factor of the PCA is composed of NO_3^-, K^+, Mg^{2+}, Ca^{2+}, Cl^-, SO_4^{2-}, Na^+, $C_2O_4^{2-}$, NH_4^+ which are positively clustered around the axis representing the main dimension 1 with 52.54% of the explanatory information of the existing relationships between the variables. The second factor (17.30%) of the principal component analysis in Korhogo is composed of $HCOO^-$, CH_3COO^- and $C_2H_5COO^-$ ions. The same interpretations as for the Abidjan site are likely valid for the Korhogo site.

In conclusion, on the three sites of the South-North transect, rainfall is strongly influenced by acidification and neutralization processes related to alkaline and acidifying species emitted by various sources (anthropogenic and natural sources). However, there are differences between the urban sites of Abidjan, Korhogo and the rural site of Lamto. At the urban sites, organic species do not participate enough in the acidification process and may not come from the same source as at the rural site of Lamto.

We performed a principal component analysis on the rainfall ion concentrations at each site for our study and came to the conclusion that this analysis showed the same findings as the correlation matrix. Therefore, since we need to reduce the manuscript size, we chose not to add the principal component analysis to the article.