

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 2#

General comments

General comment 1: The Tcarb is not described in the paper and the results are not showed.

Response to General comment 1

We have defined and added it to the table of acronyms. Results of tcarb calculations are in the table A1 in appendices and in Figure S2. tcarb is now defined as followed: the total carbonates species, calculated from this equation $tcarb = 10^{(pH-5.505)}$ (Kulshrestha et al., 2003).

General comment 2: The marine and the crustal contribution are calculated assuming that 100% of Cl⁻ and Ca²⁺ are marine and crustal. But anthropogenic sources of Cl⁻ (biomass combustion KCl) and Ca²⁺ (cement production and road resuspension) are discussed in the paper. Isn't it possible to have an estimation of their anthropogenic contributions?

Response to General comment 2

We assumed that Cl⁻ is almost 100 % from marine source because The production of sea-salt aerosol by wind stress at the ocean surface dominates the global emission flux of particulate Cl and of total inorganic Cl; on a global scale, other sources are relatively insignificant (Keene et al., 1999). But it is true that a some part could come from others sources such biomass combustion KCl, but we assume here that it can be neglected.

For Ca²⁺, we assume that it is 100% of terrigenous origin but we should make the difference between the natural origin and the anthropogenic source. Calcium naturally comes from the erosion of rocks rich in calcium. It is therefore a natural element constitutive of eroded materials (sand) or of quarry deposits. As sand and material coming from quarry deposits are widely used in construction and the production of cement, we can assume that a significant fraction of Ca²⁺ could come from anthropogenic activities. So, when we mention that calcium is 100% from crustal origin, as we refer to the sources here, we are talking about any process, either natural or anthropogenic, leading to the emission of Ca²⁺. However, we cannot at this stage distinguish natural vs. anthropogenic crustal Ca²⁺. To do it, we need additional gas phase and particulate matter measurements and also transport chemistry model data as well as detailed emission inventories for our three sites. for the moment, we do not have these data that is the reason we have just specified that terrigenous contribution in urban areas in this study is a mixture of

anthropogenic sources and crustal sources. Whereas this terrigenous contribution in rural areas is a mixture of biomass burning sources and crustal sources.

General comment 3: Data values equal to 0.000 should be replaced by the limit of detection. The authors should revise carefully the values given in the text (e.g. there are very often errors in the values).

Response to General comment 3

We have fixed this problem in revising all values given in the text and replaced 0.00 by 'limit of detection (LOD)' in table A1 in appendices.

General comment 4: The authors should rewrite several parts of the paper, and the conclusion should be clearer on the main findings (e.g. no conclusion was given on the dry/wet seasons or the impact of airmasses on rainwater composition)

Response to General comment 3

We have carefully revised all the document and rewrite some parts, especially the conclusion and the abstract in order to better highlight main findings.

Minor comments review#2

Line 22: what is a climate zone?

Climate zone has been removed from the abstract to focus more on the geographical location of sites. The notion of climatic zones is now detailed in section 3 (results) to present the climatology of each site. Now line 258-264

Lines 91-92: Arrange the references in chronological order by date of publication **Done** Now line 95-96

Lines 112-114: repetition of sentences in lines 105-107

See the same comment from reviewer 1 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.

Figure 1: unreadable text for the map a) – missing word c)

Figure 1 has been modified done

Lines 139- 143: to be shorten

In the aim to globally shorten the paper, we decided that information's given lines 139-143 were not absolutely necessary and these lines have been removed.

Line 150-151: what is the contribution of the different emission sources?

In the article of (Keita et al., 2018), percentages of contributions are given for regional (West Africa) and particularly in Côte d'Ivoire. For example: The regional contribution from each sector is presented in Fig. 4. The residential sector contributes to more than 50 % of BC emissions in Eastern and Western Africa. Waste burning is the second largest source of BC emissions in Eastern and Western Africa. For NO_x, the traffic sector is the largest contributor in Western with 30 % of the total emissions, respectively (Fig. 4b). In Côte d'Ivoire, the residential sector is the most important (58 %), followed by waste burning (26 % with 16 % residentially and 10 % in dump locations), traffic (9 %), other sectors (5 %), industry (1 %) and energy (0.1 %), and I added reference of (Keita et 2018) in this line which move to line XXX

Line 161: delete "ha" after "2000" **done, move to line 154**

Line 164: add "before "Taxi-motos" **done, move to line 156**

Line 168: please define the V Baoule, **done (now line 159)**

We wrote: "...at the tip of the "V Baoule" which represents an ecological zone of transition between the forest and the savanna".

Line 176: delete "is" before "influenced", **Done**

Line 181: remove capital letter "These" **Done**

We have deleted this part of document

Line 187-192: Is the rain gauge collocated with the rainwater sampler at Abidjan?

Yes, the rain gauge is collocated with the rain sampler at Abidjan (added in the text)

2.2 climatology: technical details are missing for the rainfall data at the Korhogo site

We move results of this section in the result section 3. The description of meteorological parameters is kept in Sect. 2.2. We specified in this section that rainfall data for Abidjan and Korhogo were provided by SODEXAM, the company that collects weather and climate data for the entire country.

Figure 2: for a better representation should add -1 and 1 in the fig (in dotted line for eg.) **Done**

Line 224: replace June by May **Done**

Line 225: replace September by October **Done**

Line 243 – 251: revise the maximum and minimum temperature (e.g., Line 243 30.83, max in fig 2 seems to be around 29.5 in March). **Done**

Indeed, this was a typo, in addition this part was moved to the results and discussion part we have revised it while trying to reduce this part now entitled "Climatology sites" in order to meet one of the concerns which is to reduce the paper. This part in move now in line 305 - 309:

"Annual mean air temperature and relative humidity over the study period in the transect Abidjan -Lamto-Korhogo are respectively $27.30\text{ }^{\circ}\text{C} \pm 1.10$ and $80\% \pm 3.89$, $28.98\text{ }^{\circ}\text{C} \pm 1.10$ and $77,05\% \pm 5.53$ and $27.00\text{ }^{\circ}\text{C} \pm 0.08$ and $60\% \pm 0.81$ (Figure 2a,2b,2c). Temperature ranged from $25.3\pm 0.20\text{ }^{\circ}\text{C}$ in august to $28.65\pm 1.85\text{ }^{\circ}\text{C}$ in march, from $27.15\text{ }^{\circ}\text{C} \pm 0.21$ in august to $31.48\pm 0.25\text{ }^{\circ}\text{C}$ in february and from $25.2 \pm 0.30\text{ }^{\circ}\text{C}$ in August to $(29.50 \pm 0.26\text{ }^{\circ}\text{C})$ in April respectively in Abidjan, Lamto and Korhogo. (Figure 2a,2b,2c)."

Line 258 – 268: revise the number of years of surplus and deficit (e.g., Line 259 from 1980 to 2020 we have 41 years but not 20+18). **Done we have revised the numbers and this part are moved in line 310 -330:**

"According to the classification of (Sarr, 2009), in Abidjan, over the 41 years, 20 years are in excess while 18 years are in deficit and 3 years have values close to the mean rainfall ($1522\pm 518\text{ mm}$). The studied period (AII) index analysis shows that 2020 (AII = + 0.14) is a

moderately wet year while 2018 (AII = -0.08) and 2019 (AII = -0.31) are moderately dry years. In Lamto, over the 23 years, 8 years are in excess while 13 years are in deficit and 2 years have values close to the mean rainfall (1229 ± 165 mm). The studied period (AII) index analysis revealed that 2019 (AII = +1.7) can be considered as a strongly wet period while 2018 (AII = -0.8) and 2020 (AII = -0.8) are classified as moderately dry years. Finally, in Korhogo, over the 31 years, 12 years are in excess whereas 18 years are in deficit compared to the mean rainfall of 1187 mm and 1 years have a value close to the mean rainfall ($1187 \text{ mm} \pm 179$). The studied period (AII) index analysis revealed that all three years are in deficit, with (AII) index values of -0.14, -0.15, -0.58 respectively and can be considered as moderately dry periods.”

Table 1: replace "inter annual" by "interannual" Done

Line 284: "INDAAF" already define line 79 Done

Line 289: add the collection surface of the sampler Done

For the needs of reduction of the document, this part has been deleted, the reader being invited to read the article references which describe fully the procedures of collection of rain samples. But the collection surface is 225 cm^2

Line 292: what is the time delay before analysis? The time delay before analysis is 6 months maximum, all the procedures are respected so that the samples reach LAERO in Toulouse in the best conditions (added in the text).

Line 366: define "VCD" Done see answer giving to the reviewer 1

Line 389: replace "NSS" by "NSSF" Done see answer giving to the reviewer 1

Line 410: remove capital letter "The" Done

Line 418: why several arrival altitudes are used in the study? Generally, people are tending to use 500m or 0.5 PBL

We have used several altitudes because the West African monsoon system is responsible of most of the rains in West Africa. So, to better understand the atmospheric dynamics controlling this phenomenon we choose to scrutiny several arrival altitudes atmosphere dynamics below 3500 meters (11,500 feet) because it is at his height that most of rainy clouds are located.

Figure 3: poor quality Done, Figure 3 has been moved in supplementary materials, it became Figure S1 and quality has been improved

Line 495: 2017 is not included in the study period

Yes, it is true, chemical composition of rain is studied over 2018-2020. However, to describe seasonal variations and define season periods for each site, we have included 2017 because of seasons overlapping over two consecutive years. In order to include the whole dry season that starts in December 2017 and ends in February 2018 we have included 2017 to generate seasonal back-trajectories

Line 557: assuming that Ca^{2+} is 100% crustal, SO_4^{2-} , Mg^{2+} , K^+ and Cl^- ratios and crustal EF are calculated assuming that Ca^{2+} is 100% crustal (Table A3)

We assume that most of the calcium would come from terrigenous sources, however, depending on the context of the study area, whether urban or rural, there are nuances. In urban areas, calcium can have several origins, while in rural areas the main source remains the terrigenous source.

Table 4: results at Abidjan, Lamto and Korhogo are not in accordance with Table A1 (eg. pH in Abidjan is 5.78 in table 4, 5.76 in table A1)

Yes, that was a typo, it has been corrected, Table 4 became Table 2.

Line 607: NSSF not shown in table 4 but in table 3

This was a typo since we have modified the paper in order to shorten it, we have moved the right table in appendices section. The new legend of this table is “Table A3”

Lines 615-637: Add references to table 3, figure 3 and so on [Done](#)

Lines 631-633: the authors indicate that air masses are “heavily loaded with dust particles” but back trajectories calculated from the HYSPLIT model just help you to understand the transport pathways of air masses, there are no evidences on the time and space aerosol load. [Indeed, there is no evidence of aerosol load. However, because the transition period \(March-April\) between the dry season and the rainy season, during which the back-trajectories showed that air masses originated globally from the Sahara and recorded highest calcium peaks, it is a period recognized in West Africa monsoon system as the period when the ITCZ is at its southernmost position, favoring the harmattan dusty air masses which are generally loaded with calcium. Therefore, we have based our conclusion on this deduction. several authors have pointed this out in their works \(Laouali et al., 2012; Marticorena et al., 2010\).](#)

Line 731: replace 0.53 by 0.58 find in Line 580 [Done](#)

Line 754: the authors indicate that “the values of nitrogen wet deposition remain lower than the critical load, estimated to be $10 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ ”. The concept of critical load defines the exposure of an ecosystem to acidification, eutrophication. Please precise if you assess the critical load exceedances for acidity or for eutrophication, and detail also how the critical load was calculated.

[We calculate these critical loads for the eutrophication phenomenon, the calculation method has been presented in these studies \(Bobbink et al., 2010; Vries et al., 2010\)](#)

Line 767-783: Do you have studied the origins of the air masses for the more acidic rains?

[No, we have not done that we should consider this aspect.](#)

Line 791 and 792: Table 5 doesn't exist [Done this was a typo.](#)

Line 795: the organic contribution to acidity is defined using sulphate and nitrate. But line 789 sulfuric and nitric acids are parts of mineral acids. It is confusing.

[That is true, it was a typo and we delete sulfate and nitrate. Move to line 656](#)

Line 803: add “organic” in “the high acidity contribution” [Done, move to line 664](#)

Figure 8: what is “Ht” in the legend?

[Ht represents rain depth \(mm\) Figure 8 became Figure S2](#)

Figure 9: add blue dots for site location [Done](#) Figure 9 became Figure 7 we add a legend

Line 1005: suppress “in Korhogo”, this [part belongs to the section2.3 which has been deleted in order to reduce the paper.](#)

Line 1052: replace 5.54 by 5.57 (see table A1) [Done, move to line 717](#)

table A4: add INDAAF in the list of acronyms [Done, move to Appendices](#)