**REVIEW OF Hagninou Elagnon Venance Donnou et al.: "Measurement report: Long-term measurements of ozone concentrations in semi-natural African ecosystems"**

**SUMMARY –** This paper analyzes 26 years of trace gas measurements (1995-2020) collected by passive filters from 14 nonurban stations across Africa, mostly in west Africa south to South Africa in the International Network to study Deposition and Atmospheric chemistry in Africa (INDAAF). There are three sections to the paper. (1) climatology of monthly ozone data from the 14 stations, organized by 4 general ecosystem types. (2) Seasonality is linked to chemical and meteorological parameters- BVOC, combustion VOC (industrial, biomass fires) and NO emissions, humidity, precipitation. (3) Trends in ozone over the period 1995-2020 are computed by typical statistical methods.

**OVERALL RECOMMENDATION** – The goals of the paper encompass a large range of topics which are too much for a single article because several aspects of the study show incomplete analyses. Of the three topics listed above, the correlations with NO and VOC (Topic 2) may be the most novel. However, the origins of those "values" and "trends" raise more questions than can be answered in one article. This Reviewer recommends dropping that material (Sections specified below) from a revised submittal and focusing on two topics: (1) ozone climatology and the relationship of seasonality and variability with the wet/dry meteorological variables and the LAI; (2) the trends and comparison with several new trend studies that include African data.

The Introduction needs to be revised to briefly review prior work that is most relevant to the INDAAF project. Here is the best place to include reference to the many campaigns held in west and southern Africa (Listed in 3c below) that examine processes, particularly related to fire impacts on ozone, but that are only "snapshots in time" compared to the long-term INDAAF measurements. The authors should put some of their existing references to meteorological and seasonal influences in the Introduction to give context and to motivate the reader to understand how their seasonal analyses (Section 3.1) extend and give new insights into the earlier work. (The goal here is to highlight WHY your study is original and important!)

In revising subsequent Sections: (1) For the <u>ozone</u> <u>climatology</u>, a priority request is more detail about quality control in passive sampler datasets. (2) For <u>ozone trends</u>, the results shown (Figure 15) should be augmented with trends to display all 14 sites (Section 3.5). Additional context for interpretating these results is needed, including comparisons with other publications on African ozone trends. How do the trends over South Africa compare, for example, with other studies? Similar, different, why? <u>In summary, the Reviewer recommends the paper for ACP publication *after* major revision. The latter must include better evidence of INDAAF ozone data quality and more thorough analyses of the 1995-2020 trends at the 14 stations.</u>

"Measurement Report" does not sound like an appropriate classification for a paper that presents more original scientific results than simply documenting the existence and archiving of a dataset. The reviewer suggests dropping the "Measurement Report" heading. A more suitable title might be: "Surface ozone seasonality and trends over Africa (1995-2020) from the INDAAF project".

**MAIN COMMENTS**

1. <u>Abstract.</u> A major concern about the ozone measurements is quality control of the passive sampler data (more below). A sentence or two that documents the reliability of the INDAAF station ozone observations is needed in the Abstract. Lines 33-34 – Sentence is ambiguous as written. Do you mean southern Africa has higher mean ozone concentrations or greater seasonal variations in ozone in east Africa? The Sahel region sites are not defined although it is recommended that the two sentences in Lines 34 to 37 be omitted. Line 40 –

specify the dates of the trend calculations. The lines 42 to 44 refer to changes in VOC and NO2 that appear to be assumptions. That is, the paper gives no measurements to support this statement for these 2 INDAAF sites. These lines should be removed.

2. Sections on NO, VOC, BVOC. Three reasons for *recommending nearly all the material in Sections 2.4, 3.3, and 3.4 be removed from the current paper*. (a) The paper is too long and yet, important information is still lacking. Example: explanation of the method in 3.4.1 needs to be expanded. (b) It is not clear to what extent values for the species NOx, VOC, BVOC, their seasonality of emissions, changes over time, are from model(s) and/or experiments and if so, which ones? Explain. It is important when referring to published relationships between ozone and the precursors to distinguish between links based on measurements (clarify with references to the data sources and publications) and "trends" based on models that are of unknown accuracy. Related to this point, the relationships for species and ozone in Figure 12 are not convincing, in contrast to those in Figure 2 where there are definite links of ozone with climatic/meteorological parameters. (c) The correlations in Figure 14 and Table 5 are intriguing but not very clear. For example, previous studies demonstrate biomass fire impacts on sites in northeast South Africa but how is the attribution to VOC and NO combustion made here? *In summary, the Reviewer finds good potential in linking ozone with the precursors and processes (biogenic vs combustion NOx and VOC, for example) but the analyses in this paper are confusing in some places and are not convincing in other sections. Recommend that the authors move these analyses to a second paper that presents ozone and precursor relationships with more rigor and *observational* evidence.*

3. There are a number of places where statements should be made more accurately. Many suitable references are given in the paper but sometimes in a misleading way. Examples from **Section 1 follow**:

a. Line 61. Since Zhang et al. (2016) both modeling and observational studies have shown that ozone trends are not uniform regionally or seasonally, i.e. even in the tropics a number of sites with ozonesonde profiles exhibit no trend (Thompson et al., 2021). A study with sondes over equatorial southeast Asia, published in the TOAR II collection by Stauffer et al. (2024), shows no definite ozone trend annually but a 6-8%/decade increase limited to 3 months/year. *Insert words to this effect on Line 62 after Adon et al., 2010.* The references are here:

Thompson, A. M., R. M. Stauffer, K. Wargan, J. C. Witte, D. E. Kollonige, J. R. Ziemke, Regional and seasonal trends in tropical ozone from SHADOZ profiles: Reference for models and satellite products, J. Geophys. Res., https://agupubs.onlinelibrary.wiley.com/doi/10.1029/2021JD034691, 2021.

Stauffer, R. M., A. M. Thompson, D. E. Kollonige, N. Komala, H. Khirzin Al-Ghazali, D. Y. Risdianto, A. Dindang, A. F. bin Jamaluddin, M. Kumar Sammathuria, N. Binti Zakaria, B. J. Johnson, P. D. Cullis, Dynamical drivers of free-tropospheric ozone increases over equatorial Southeast Asia, Atmos. Chem. Phys., https://doi.org/10.5194/acp-24-5221-2024

b. Line 63-65. Laban et al. (2018), which describes patterns of South African ozone at a number of sites that are both rural and anthropogenically influenced, emphasizes that the highest ozone is in the dry season in the semi-arid areas in the Highveld, largely due to a maximum in domestic fuel usage for winter heating, not biosphere interactions, as the paper states.

c. Lines 70-75. The Reviewer is not sure if the continents of South America and Africa were compared that Africa (at least sub-Saharan Africa) is "the least studied." On line 73 it is more accurate to say "*may be* the least studied continent." Over the past ~30 years there have been many large African field campaigns conducted (AMMA, EXPRESSO, SAFARI/TRACE-A, ORACLES, SAFARI-2000) such that hundreds of articles have been published on African air quality and environment. Links to dynamical factors affecting ozone seasonality (Diab et al., 1996; 2003; 2004), interannual variability in ozone related to ENSO (Balashov et al., 2014) and widespread impact of biomass and domestic fires in southern Africa are well-established. On the latter, refer to detailed analyses of ozone and related measurements in Special Issues of *J. Geophysical Res.* on SAFARI/TRACE-A, and SAFARI-2000. Southern Africa has been the major arena of these ozone studies and South Africa has had several high-quality monitoring programs. In Line 65 it is more accurate to say "With the exception of South Africa, ozone variability is poorly documented on the African continent." For example, in North American and European journals, North Africa probably has the smallest number of articles. However, having said that there are many African studies about ozone and air quality, the number of measurements *publicly available* may be very small. The authors can point out that the INDAAF is among the few datasets that are available to the scientific community. Diab et al: Diab, R. D., A. M. Thompson, M. Zunckel, G. J. R. Coetzee, J. B. Combrink, G. E. Bodeker, J. Fishman, F. Sokolic, D. P. McNamara, C. B. Archer, and D. Nganga, Vertical ozone distribution over southern Africa and adjacent oceans during SAFARI-92, *J. Geophys. Res.*, 101, 23,809-23,821, 1996; Diab, R. D., A. Raghunandran, A. M. Thompson, V. Thouret, Classification of tropospheric ozone profiles over Johannesburg based on MOZAIC aircraft data, Atmos. Chem. Phys., 3, 713-723, 2003; Diab, R. D., A. M. Thompson, K. Mari, L. Ramsay, G. J. R. Coetzee, Tropospheric Ozone Climatology over Irene, South Africa from 1990-1994 and 1998-2002, J, Geophys. Res., 109, D20, D20301, doi: 10.1029/2004JD004293, 2004.

d. Lines 100-105. A number of references are given about studies and campaigns but little information about the findings of each that are relevant to the authors' study. What the reader wants to see is "what do we know from the prior campaigns?" "Do they agree with one another?" "What are new INDAAF results that confirm, contradict or complement the earlier findings?" The list in these lines is not useful without connecting the background to the current paper.

e. In like manner to comment (b) above about Line 65, LINE 106 is not correct. There have been a number of studies with South African ozone and related data. Line 106 should say "With the exception of South Africa very little information is available on the long-term evolution of O3 chemistry over Africa. The impact of meteorological parameters and atmospheric chemistry… and the analysis of long-term trends *is only partially explained.*" *In other words it is not correct to say the trends are "unexplored*." The authors cite Balashov et al. (2014) (line 537) which determines trends for 5 South African sites with high quality surface O3 data, over ~15 years; see also Martins et al, 2007. Trends over a longer period, ~1990 to 2011 in ACP, described the seasonality of free tropospheric O3 trends over Irene [Pretoria], South Africa with both IAGOS and ozonesonde data (Thompson et al., Atmos. Chem. Phys., **14,** 9855-9869, 2014; the latter paper corrected a sampling error in Clain et al., 2009). In ACP, Gaudel et al. (egusphere-2023-3095) use IAGOS African aircraft data to estimate trends over a number of sub-Saharan African cities for the period ~1995-2019.

4. Section 2.2. This section, fundamental to the quality of the paper, is inadequate. It is not enough to cite previous papers to establish the accuracy of the INDAAF ozone record. Uncertainties in ozone mixing ratios for typical samples need to be provided. Uncertainties are also needed in the graph of trends in Figure 15. To give further confidence in the ozone time series for the INDAAF sites, comparisons of sampler ozone with independent ozone measurements should be made for sites where the latter data are available within the 1995-2020 period. Examples: ozone from Irene, South Africa, sondes at the surface and Welgegund (continuous ozone monitor) can be compared to LT; if ozone from the WMO/GAW station at Cape Point is available from an independent analyzer, a comparison of trends from such an instrument during the INDAAF period should be made. Although Nairobi observations are not co-located with the Mbita INDAAF data, their ozone should display similar seasonal patterns. Nairobi ozonesonde data are available from SHADOZ (1998-2023; https://tropo.gsfc.nasa.gov/ shadoz); surface monitor ozone data may also be available.

5. Sections 3.1 and 3.2.

The analyses corresponding to Figures 3-9 (with Table 3) are very good. In later Sections, e.g., 3.4.2.3, there are references to meteorological influences on South African ozone seasonality (humidity, temperature; Balashov et al., 2014; Laban et al., 2018; 2020). The Introduction and/or earlier sections would be strengthened by moving some of these references forward.

Figures 4, 6, 8. These Figures originate from averaging the values in Figures 3, 5, 7, respectively, no? Please explain in the captions.

Figure 9. Likewise, although parts of the text define the periods of “wet”, “dry” season, it would be useful to repeat or summarize those definitions in the Figure 9 caption.

Page 12. Discussion. References to Nepal climatology are not relevant here. Remove.

Pages 16 and 17. These comparisons are not relevant to the paper. There is no value to Table 4 and it should be deleted along with Lines 380-391. We don't know that the same analytical methods are used as at INDAAF sites. The dates are not a match for the INDAAF period in many cases._What is the point of making comparisons with Arctic, Antarctic, midlatitude sites?

Figure 10. This Figure *is* relevant to the discussion of INDAAF ozone climatology. In particular, the INDAAF data for South Africa should be added to the Figure and those values should be compared to earlier publications with South African data. Reasons for similarities and differences should be discussed, including how sampling systems for the non-INDAAF data compare to INDAAF measurements in similar parts of South Africa.

6. Sections 3.3, 3.4

As mentioned above, most of this material should be removed or saved for a separate manuscript because it is about precursor variability and trends for which necessary explanations, references and background are beyond the scope of this paper.

Section 3.5

The focus on O3 trends is one of two major results of this paper but the discussion is incomplete (no South African data displayed, for example). Some points raised in Section 3.5.1 refer to BVOC trends (? From models; if so give short explanation). There have been trends papers with African ozone data that cover most of the INDAAF period analyzed here, ~mid-late 1990s through 2020: Gaudel et al., 2018; 2020; in review- egusphere-2023-3095; Thompson et al., 2021. How do the authors' INDAAF trends agree? Discuss in more depth. There should be references on seasonality of fire impacts from the Piketh and van Zyl-Beukes groups. Expand the literature search. It may be possible to add truly relevant articles to the bibliography and remove references on biogenic emissions or model “trends” that are not data.

**Miscellaneous Comments and Questions:**

**Abstract.** Place names. Location, Country; - separate with semi-colon. Banizoumbou, Niger; Katibougou and Agoufou, Mali; etc

**Line 53** Don't begin sentence with O3, use “Ozone”

**Line 94.** “Previous studies” – delete “existing”

**Line 112**. Delete the 2$^{nd}$ occurrence of “first”

**Lines 116-120**. Recommend omitting 3 sentences. *A second objective is then: “In the 2$^{nd}$ objective, we use non-parametric statistical tests to assess seasonal and annual trends in O3 in the context of other trend analyses of Africa ozone (Thompson et al., 2014; Gaudel et al., 2018; Gaudel et al., 2024).”* Option to add: ”In a companion/separate article INDAAF ozone trends are linked to potential changes in NO2, VOC, BVOC.”

**Table 2**. For South Africa LT, Sk, Af – End point is 2015. Did the program end there or are there data taken since 2015 that are not publicly available?