

Review of the manuscript “The white-light humidified optical particle spectrometer (WHOPS) – a novel airborne system to characterize aerosol hygroscopicity”, by B. Rosati, G. Wehrle, P. Zieger, M. Gysel, U. Baltensperger, and E. Weingartner.

The paper describes the development of a system to retrieve the real part of the complex refractive index (RI) of aerosols, and characterize their hygroscopicity, i.e., the growth factor (GF). The system utilizes a DMA-humidifier-white light optical particle counter (WELAS) to retrieve the two variables. The manuscript is well written and has a very nice laboratory validation of their system, it is thorough and addresses all the caveats the WELAS has. However, I do not think the validation done in the laboratory translates directly to the field measurements. The field measurements are a crucial part of the paper, as indicated both in the title, and in the discussion. However, the field measurement data presented in the paper requires major revision before it can be published. Since the main focus of the paper is its weakest point at this stage, I do not recommend publication in the present form. Following are my main concerns, which should be addressed before the manuscript is suitable for publication.

Regarding the retrieval of the RI; The retrieval of the RI in the laboratory is done by size selecting several diameters, an RI is retrieved for every size, and then the average RI of all the sizes is taken as the final retrieved RI and compared to literature data. For PSL, four different sets of measurements were done with a mean $RI = 1.60 (\pm 0.04)$, but if you look specifically at the RIs from a single diameter, for example 500nm, the RI varies from 1.55 to 1.6 for the two data points shown. For a 300nm diameter the RI varies from 1.53 to ~1.625. These variations are larger than the stated error. This RI is then taken as an input variable to obtain the GF. The authors do not show how these variations for retrievals done with a single diameter are reflected in the GF calculations, specifically of the error. The authors even mention in page 7335 line 10-11: “Figure 4 demonstrates the **crucial** importance of using the **appropriate index of refraction** for the optical sizing of grown particles...” Also, what is the convergence criterion for the RI retrieval using a single diameter? In their look up table(s) they look for a dry RI corresponding to a dry cross section and a $GF=1$, what is the interval between the ‘steps’ of the RI? In other words, do they vary the RI by 0.01 or 0.001? Also, can the authors state what wavelength range is the RI retrieval valid for?

I also think the field measurement analysis of the Zepelin campaign is missing important data to help the authors validate their proposed airborne system. The authors mentioned they had an Aerosol mass spectrometer (AMS), a Scanning mobility particle sizer (SMPS), an anemometer, meteorological data from the zeppelin, and another WELAS to help validate their new system, but the data of these instruments was not used at all. The authors should incorporate the data from the additional instrumentation. The authors mentioned that a change in wind direction “obviously” demonstrates a change in air mass. I think the authors should show a full meteorological analysis to support this claim.

They can show wind speed, temperature, RH, particle concentration, and size distributions from the SMPS. If wind direction alone can indicate a change in air mass then the changes seen at the beginning of the campaign, between 14:55 and 15:30, will also have to be changes of air mass. Fig.11c should have wind speed in the left y-axis and the wind direction can be depicted with arrows, with a separate arrow showing what direction signifies north.

The authors mentioned that Fig. 11d shows the measured mean GF and kappa values (black line) for 500nm particles. The authors need to show the error bars. If what the authors define as Part 1 is the same as mass, how do they explain the large variations from a mean GF ~ 1.7 to a mean GF ~2.6? How can a variation from a GF~2.3 to a GF~3 occur in what looks as two consecutive measurements (See Fig. 11b at 51.9N – 4.6E)? I think the SMPS and AMS data can help resolve this issue, by providing the concentration and chemical composition of the particles (and specifically the concentration of the 500nm particle), respectively. Also, the authors should better explain the purpose of adding mean growth factor of particles larger than 1.5 in Fig. 11d (red and green lines).

The authors mention in page 7345 line 15-18 that they did back trajectory analysis, but that it is not possible to assign air mass types to the two parts they define. But the authors could define air mass type for certain measurements. For example, for the measurements taken at around latitude 52.5N – 3.9E, a back trajectory analysis can be done; see Fig. 1 below.

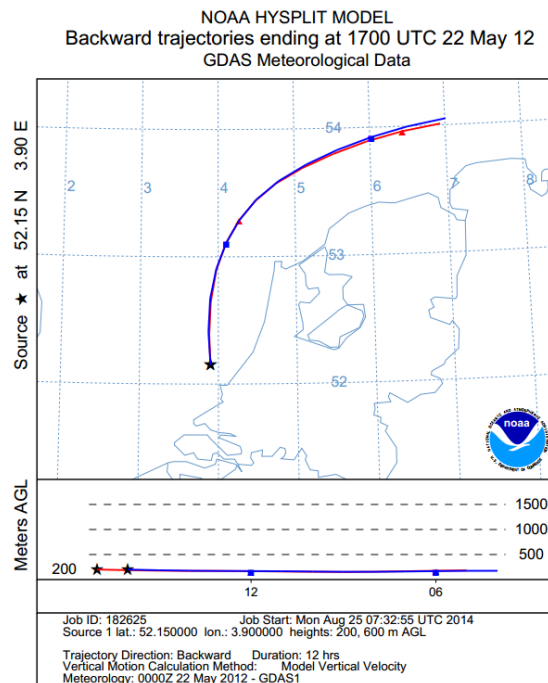


Figure 1Review. Back trajectories (12 h) arriving at 17:00 for height levels of 200m and 600m.

Fig. 1Review shows that at both levels, 200m and 600m, the air mass arriving at the measurement point had been at least 12h above the sea. Hence, sea spray aerosol was most likely measured. The same can be done for the aerosols that were measured at the beginning of the campaign:

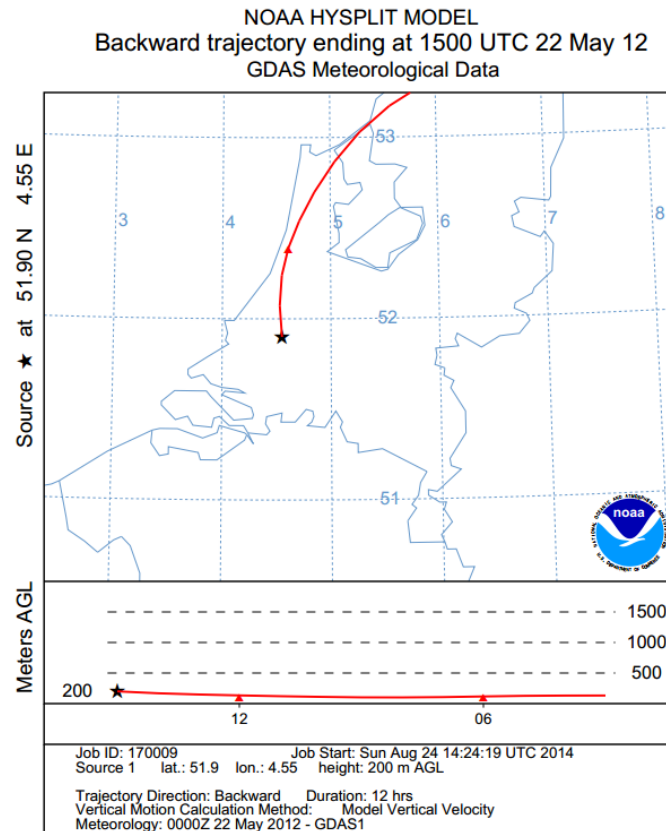


Figure 2Review. Back trajectory arriving at 15:00 for height level 200m

From Fig.2Review, the aerosols that were measured at 15:00 (the beginning of the campaign) had more continental influence than the ones measured at 17:00. From Fig. 11b in the paper, the GFs measured at the two points shown in Fig1Review and Fig.2Review have approximately the same value $GF \sim 1.8$, can the authors explain this? This quick analysis can of course be greatly improved (e.g., using the AMS, SMPS, wind speed and direction, etc.), but I think the authors need to show that the results given by the WHOPS are consistent.

The authors mention that on average 15% of the particles had $GF < 1.1$, which could be explained with externally mixed dust, soot, or biological particles. Here, for example, they could use the aethelometer data to verify the influence or non-influence of soot.

What was the average number of particles measured to obtain the RI and GF for the two diameters used? Why do the authors use a log scale in the x-axis of Fig.11e and Fig.11f? Also, why is $dN/d\log GF$ used? Are the widths of the GF bins different? What are the widths? If they are, this should be mentioned in the method section, and explain why there is a need for different widths in the GF measurements.

The authors mentioned that they found a rather constant RI for the whole campaign, with a value of 1.42 (± 0.04). However, they found a great variability in the GF. The variability in the GFs would imply different type of particles; wouldn't the authors expect a different RI as well? It is strange to have such a big variability in GF but a constant RI, can the authors elaborate on why the RI remained constant?

Would high concentrations of NO_x or O₃ affect the measured diameter in the WELAS? Were high concentrations of NO_x and/or O₃ encountered during the campaign?

Other comments:

-Was the counting efficiency experiment done with the 0.5lpm WELAS or the 5.0lpm WELAS? Would the nozzle that the company inserted make a difference?

-When deriving the RI from a single diameter, was $D_{dry,mob}$ assumed to be one diameter, or was a narrow size distribution used? Is there a difference in the retrieved RI?

-I don't think the authors should use the qualifiers: "more hygroscopic", "non- or slightly hygroscopic", etc. It is ambiguous. They can just say 74% of the particles had GF > 1.5, for example.

-Page 7325, line 20: Add the country to the WELAS description

-Page 7327, line 16: Add company, city, model, country, etc.

-Page 7328: What is PROMO?

-Page 7332, lines 18-20: Hand and Kreidenweis (2002) should be added, they also have a similar approach

-Page 7342, line 7: 'vales' should be 'values'

-Page 7344, line 13: change 'nm' to 'm'

-Page 7345, line 6: 'Fig. 11c' should be 'Fig. 11d'. Line 19: 'Fig. 11d' should be 'Fig. 11e'

- Page7364, Fig.10a. The legend is wrong. 'ADDEM' is shown for the measured GF.

- Page 7365-7366. Why does Fig. 11 have two captions? Maybe it will be better to put the 'second' Fig. 11 as Fig. 12?

Reference:

Hand, J.L., Kreidenwies, S.M, A new method for retrieving particle refractive index and effective density from aerosol size distribution data, *Aerosol Science and Technology* 36: 1012–1026, 2002.