

***Interactive comment on “Aerosol properties associated with air masses arriving into the North East Atlantic during the 2008 Mace Head EUCAARI intensive observing period: an overview” by M. Dall’Osto et al.***

**M. Dall’Osto et al.**

colin.odowd@nuigalway.ie

Received and published: 30 March 2010

Atmospheric Chemistry and Physics Discussions

Interactive comment on “Aerosol properties associated with air masses arriving into the North East Atlantic during the 2008 Mace Head EUCAARI intensive observing period: an overview” by M. Dall’Osto et al.

Anonymous Referee #1 Received and published: 24 February 2010 Review of "Aerosol properties associated with air masses arriving into the North East Atlantic during the

C11831

2008 Mace Head EUCAARI intensive observing period: an overview" by Dall’Osto et al.

The manuscript presents a long and somewhat meandering overview of the intensive measurements carried out at Mace Head as part of the EUCAARI project. The paper necessarily presents a comprehensive characterisation of the climatology experienced during the project and sets up the scene for a series of more detailed papers. Whilst it is difficult to determine great scientific merit in the current work in its own right, such an overview provides the background needed for subsequent work. Given the nature of the paper, the authors are to be congratulated on keeping it readable. I find the paper is suitable for publication in ACP once the following detailed, largely minor, comments are addressed.

We appreciate the reviewer’s time and constructive comments.

Abstract: Attribution of the nucleation mode in the Arctic air to open ocean nucleation is at best ambiguous. "Black carbon concentrations in polluted air were between 300–400 ng m<sup>-3</sup>, and in clean marine air were less than 50 ng m<sup>-3</sup>" - this is an operational definition, not a conclusion and should therefore not be in the abstract. Well, it is also based on air mass trajectories – nevertheless we have removed it from the abstract.

There is a problem with the statement: "Although the concentrations and size distribution spectral shape were almost identical for the young and aged continental cases, hygroscopic growth factors (GF) and cloud condensation nuclei (CCN) to total condensation nuclei (CN) concentration ratios were significantly less in the younger pollution plume, indicating a more oxidized organic component to the aged continental plume."

Whilst the conditions stated are necessary for the concluded statement, they are insufficient. It is possible that any differences could be attributable to differences in the mixing state between the young and aged populations. If it can be stated that the mixing states were also similar, then the statement is more likely true. However, it is then difficult to understand why aerosol in younger air is not more externally-mixed than in

C11832

aged air. Is it not possible to more directly deduce that the aerosol in more aged air-masses is more oxidised, such as through the HR-ToF hi-res analysis? It is, after all, not a very surprising finding.

There is evidence in GFs for more external mixing in the younger air mass. Additionally, the HR AMS data shows a higher degree of complexity in the organic component for the younger air masses, with more PMF factors including Hydrocarbon-like aerosol (HOA) and more fresh Secondary Organic Aerosol (SOA) (manuscript in preparation).

1. Introduction: p26268, line 8, sentence beginning "In terms of climate..." is confused and has inappropriate parentheses. Corrected p26268, line 20, "Acquisition" to be corrected Corrected

p26269, line 14, "An 12 months..." to be corrected Corrected

p26269, line 27, O'Connor et al., 2009, reference not found Corrected to 2008 p26270, line 6, replace "humidity" with "hygroscopicity" Corrected

p26271, line 4, "Micrometeorological" to be corrected Corrected

p26271, line 17, "mode" to be corrected Corrected

p26272, line 15, 16.1 or 16:1? Corrected

p26274, line 4, How well do these values agree with theory? Which theory? Is this agreement expected / good enough? Text added: "and were found to be within a growth factor deviation of  $\pm 0.05$  compared to previous recorded data (Tang, I. N. and Munkelwitz, 1994)."

2. Methods: Overall a good enough description of all techniques, with the right balance of detailed description and references to previous publications.

#### 2.4 Aerosol Mass Spectrometry

p26274, line 5, Title to be corrected Corrected

C11833

p26275, start line 4, It is stated that the MS mode was run in V or W mode. It should be stated in what mode the pToF data was collected as size-resolved Hi-Res data is a valuable potential product, but very thin on the ground! The following text is already in the manuscript.. The AMS was run in "V-mode" with a mass resolution of up to 3000  $m/z$  alternatively with a "W-mode" by using a second reflectron, which increases resolution to 6000  $m/z$  but decreases sensitivity by approximately one order of magnitude.

3. Meteorology: p26278, line 23, Which is it - "Marine Tropical" or "Marine Sub-tropical"? Reference to the wrong classification should be deleted. Sub-tropical is not an official air mass classification so tropical is kept with the qualifier that sub-tropical is perhaps a better description.

3.2 Aerosol concentrations size distributions p26280, line 8 A lognormal mode has no intrinsic physical significance and is merely a mathematical construct. The last phrase of the sentence should be rephrased: "...4 modes in log concentration space most readily represent the size distributions or "... the size distributions require a minimum of 4 log concentration modes to adequately represent them". We are not talking about size distributions here – it is the frequency of concentration occurrence distribution. No changes made.

p26281, line 3 and figure 7 - it's somewhat surprising at first glance that the total number concentration during nucleation events is, in all cases, lower than during nonnucleating periods.

p26282, line 2 - I presume you mean figure 7b here, not figure 6. If so, the highest period-averaged number concentration refers to that in Figure 7a. It is evident that 7b has the nucleation events since the size distributions are open - but as I mentioned in my last point, it appears peculiar that size distributions with coastal nucleation have lower number than those with. The 3054 referring to the lowest period number concentration is also that found on the "non-nucleation" plot 7a. I am confused. Text clarified: the

C11834

figure 7 a and b were backward in order - this is not changed. In the original 7b, what is presented is non-nucleation data. The "open" size distributions are simply noise due to the nSMPS; however, the noise concentration is 1-2 orders of magnitude lower than that found in the nucleation event data.

3.3.1 Online chemical composition - PM<sub>2.5</sub> mass AMS comp mass : p26283, lines 17-20. It is stated that the particles were completely neutralised during continental periods, but inferred that, because sulphate was in much greater molar excess of the ammonium, this was not the case for marine air. Obviously, the particles could have been neutralised by the unmeasured sodium, so this should be explicitly stated (noting that a "particle type internally mixed with alkali metal cations and sulphate but no nitrate" is one of those identified in the ATOFMS section in the clean marine air).

The ATOFMS indeed found a particle type internally mixed with alkali metal cations and sulphate, but unfortunately we cannot address the quantitative apportionment of the two components therefore the ATOFMS does not allow to understand the neutralization behaviors of particles sampled in clean air masses. The collection efficiency of the AMS depends on the Under typical continental conditions the AMS collection efficiency (CE) is about 50%. Typically this value is rationalized by the supposition that approximately 50% of the particles bounce off the vaporizer before they evaporate. For liquid particles and pure sulphuric acid particles, collection efficiency increases to almost 100%. The higher CE for marine samples suggests a higher degree of sulphuric acid particles, giving the similar RH sampled during the campaign. Moreover, and most importantly, off-line techniques data (section 2.5 and 3.3.2) showed that the component of non sea salt sulphate was much higher than the sum of the cations (both metals and ammonium) in the submicron marine aerosols.

p26284, line 4 kind of answers an earlier query in stating that the size-resolved AMS data are presented at unit mass resolution, but the sentence following on from this starting on line 6 about MSA is unclear and grammatically incorrect. This should be rephrased and clarified.

C11835

Rephrased: Consequently, the MSA component is split broadly between the organic fraction (67%) and the sulphate fraction (28%) of the signal (Dr. F. Drewnick, pers. comm.).

p26285, paragraph starting line 28 - it is stated that peaks at 48 and 64 (Ti and TiO) are present in the +ve spectrum, but from line 11 p 26286 it is stated that "No dust Ti-rich particles were found during EUCAARI". Please clarify.

Whilst metals like Na, K, Ca, Mg, Ti are common in ATOFMS mass spectra of dust particles, we did not find any ATOFMS mass spectra with dominance at m/z 48 and m/z 64 (so called Ti-rich particle types) such as previous field campaign at the same location (Dall'Osto et al 2004).

3.4 Hygroscopicity: It is now becoming quite clear that Mace Head receives a fair amount of organic material in its particulate loading and that these lead to significant differences in water uptake properties etc... to those received by other marine sites. Can the authors offer an explanation as to why Mace Head has more organic aerosol material than, say, in the Mauritanian upwelling where the waters are extremely nutrient rich or the North Sea where there is a significant amount of organic pollution? There are biologically richer and more polluted waters than the Atlantic, but seldom airmasses with more organic aerosol material. I think this is an important question as parameterisations of organic seaspray fluxes are being derived from the N. Atlantic.

Not at this stage – while we suspect some reasons, they would be too speculative.

---

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 26265, 2009.

C11836