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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The composition of nucleation and Aitken modes particles during coastal nucleation events: evidence for marine secondary organic contribution" *by* P. Vaattovaara et al.

P. Vaattovaara et al.

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We would like to thank the referees for their useful comments.

Major comments (referee #2)

Author comments:

Discussion on the results presented in Figure 3 associated with laboratory PHA-UCP will be rewritten, as well as lines 4-23 on the page 3343.

The purpose of that Day 7 June (p. 3347) was to demonstrate the importance of the

solar radiation intensity for new particles formation events (Figure 4). Because the importance of solar radiation is well known already earlier, Day 7 June text will be removed as recommended.

P. 3348, starting from line 14: Indeed, CCN concentrations availability has been earlier observed to increase during a nucleation event at Mace Head site (O'Dowd, Biogenic coastal aerosol production and its influence on aerosol radiative properties, Geophys. Res. 106, D2, 1545-1549, 2001). Also interestingly to this study, Aalto and Kulmala (Using a cloud condensation nuclei counter to study CCN properties and concentrations, Boreal environment research 5; 349-359, 2000) reported that 50 nm particles have 60% activation efficiency at 0.5% water supersaturation with about 0.8 water soluble fraction (compared to ammonium sulfate) in the marine air mass at the same site. Furthermore, Väkevä et al. (Hygroscopic properties of nucleation mode and Aitken mode particles during nucleation bursts and in background on west coast of Ireland, Geophys. Res., 107, D19, 2002) used CCNC (cloud condensation nuclei counter) with 0.5% water supersaturation for particle diameters from 150 nm down to 15 nm (i.e. 150, 112, 84, 63, 47, 35, 27, 20, and 15 nm) at Mace Head. The results show that with 0.5% water supersaturation the average ambient particles had to be eight nanometers larger than the ammonium sulfate particles used in the calibration in order to be activated.

The paragraph starting from page 3353 and continuing to page 3354 will be partly rewritten.

In order to take open ocean into account in the conclusions, p. 3354 lines 23-27 and p. 3355 lines 1-2 will be transformed to the following form: Additionally, our UFO-TDMA results suggest that biogenic secondary organic compounds originating from marine coast and open ocean biota may, in addition to being significant contributors to the nucleation mode processes, affect the formation (i.e. size, composition and thus, variety of properties) of CCN and even larger radiatively active particles (e.g. those produced by bubble bursting), subsequently playing an important role at least in the regional radiative budget, influencing also different feedback mechanisms between

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marine coast and open ocean biota, aerosols, clouds, and solar radiation.

Also p. 3355 lines 27-29 and p. 3356 lines 1-6 will be changed to the following form: From the wider perspective, biogenically driven secondary organic contributions can be anticipated also at other biologically active, species specific marine coast and open ocean waters (covering parts of all five oceans, especially at temperate and polar zones; see Fig. 1c and also Kulmala et al., 2004a). It is worth of noticing that the surface of biologically active open ocean regions is really large. Furthermore, similar effects could also be possible in fresh waters (e.g. lakes; see Fig. 1c). Thus, from the global perspective of particle and CCN formation and their climatically important properties, it would be crucial to experimentally investigate biogenic secondary organic contributions at those biosynthetically active ocean areas and fresh waters located around the world.

Generally, "marine/coast" type terms will be transformed to "marine coast and open ocean"

Minor/technical comments (referee #2)

Author comments:

Introduction: It is taken into account that DMS is not a sole source of non-sea salt sulfate over the northern Atlantic. Thus, the sentence starting from line 11 (p. 3340) will be moved to the following form: ..., the formation of which can originate via DMS from algae or sulfur from antropogenic origin, is only a minor...

We would prefer to keep Figure 1. Figure 1 shows how biological activity changes in different parts of the Atlantic Ocean (marine coastal and open ocean) during two measurement periods (spring and autumn 2002). The figure also shows that the ocean really is biologically active during those seasons. The Atlantic Ocean spring period seems to be more biologically active than autumn or average situation, so the biological activity of the spring period can be defined to be enhanced. It is possible to compare

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Atlantic Ocean biological activity values to the values of other oceans during spring and autumn seasons and average situation. Biological activity values describe the concentration of chlorophyll a and thus the amount of phytoplankton which is though to be very important source of organic compounds (e.g isoprene) on the oceans. Thus, Fig. 1 indicates the most potential organic source regions around the world oceans and even in the biggest lakes. Those potential organic source regions of the oceans are in good agreement with worldwide observed coastal nucleation events. Thus, the figure is important both for regional research but also for wider perspective and future research, especially at marine coast and open ocean regions.

We will combine and make changes (axis titles,scale) to Figures 7, 8, 9, 12 and 13 as required.

The manuscript will also be proof read carefully after making the final corrections.

Major comments (referee #1)

Author comments:

In the start of introduction (p. 3339, lines 1-2) it will be explained briefly what a nucleation event is: Bursts of nanometer-sized particles formed from nucleated atmospheric gases and grown to detectable sizes (i.e. nucleation events) have been observed at coastal and marine environments around the world (see Kulmala et al., 2004a).

The reasoning for the choice of particular compounds as well as the discussion of chlorinated and brominated hydrocarbons will be separated from the instrument description.

PHA-UCPC text will be rewritten including a short description of the operating principle. Figure 3 gives probably some help for non-expert to understand text.

An overview of what type of information (i.e. back trajectories, particle size distribution and particle concentration, ocean chlorophyll a concentration, black carbon data, low tide data, wind strength and direction, solar radiation, air pressure, relative humidity)

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was available for the data-analysis during these measurements will be given in the methods section.

We will not use the words obviously and naturally on the page 3345 (lines 16-25) and the Figure 4 will be explained better: The figure shows that during the strong events (days 4-6 June), the largest observed nucleation mode particles reach 10 nm. In addition, the growth rates of the freshly nucleated particles seem to be quite high. During the same campaign, O'Dowd et al. (Airborne measurements of nucleation mode particles I: coastal nucleation and growth rates, Atmos. Chem. Phys. Discuss., 6, 8097-8123, 2006) made airborne measurements of nucleation mode particles growth rates along the west coast of Ireland, in the vicinity of Mace Head. They found that close to coastal plume head ($<$ 1 km) growth rates can reach values as high as 123-171 nm h-1, decreasing gradually to 53-72 nm h-1 at 3 km. Further along the plume, the growth rates were calculated to be 17-32 nm h-1 at 10 km. Those growth rates are quite high compared to boreal forest cases, for example (see e.g. Kulmala et al., 2004a), but comparable to the earlier reported growth rates of 15-180 nm h-1 for recently formed nucleation mode particles at Mace Head (O'Dowd et al. (A dedicated study of new particle formation and fate in the coastal environment (PARFORCE): Overview and objectives and initial achievements, Geophys. Res., 107, D19, 2002) and Dal Maso et al. (Condensation, and coagulation sinks and formation of nucleation mode particles in coastal and boreal forest boundary layers, Geophys. Res., 107, 2002). The growth rates strongly imply that the largest nucleation mode particles observed not forming very far from the observation site. Thus, the measurements of sub 10 nm particles are especially important for characterizing those newly-formed particles.

The uncertainties of the measured growth factors will be mentioned on the page 3346 starting from line 26: Additionally, the GFs uncertainties (i.e. $<$ 0.02 for 6-8 nm and $<$ 0.01 for 10-50 nm; based on Vaattovaara et al., 2005) have an effect too.

P. 3347, line 23: There were 12 event days during the campaign and few were both clean and strong enough for analysis. Mainly 30 nm and 50 nm were not in the same

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mode, although their overall GF trend is similar between different event days, and actually between all days. Other events were mainly both more polluted and weaker than the events already analyzed and thus their effects on Aitken mode particles were not very clear. The best example event day to show that 50 nm particles behave similarly to the smaller Aitken mode sized particles is 22 May, when 50 nm and even 20 nm particles were in the same mode. In that case, GF of 50 nm particles increases from 1.08 to 1.11 and GF of 20 nm increases from 1.08 to 1.13 at the same time. Interestingly, Väkevä et al. (2002) noticed at Mace Head that CCNC data for 50 nm particles show similar characteristics of aerosol solubility compared with the 20 nm particle data from UFH-TDMA (ultrafine hygroscopicity tandem differential mobility analyzer). The sentence on the page 3347 lines 21-24 will be changed to the following form: However, it is likely that 50 nm and particles behave similarly to 30 nm particles, because 30 nm and 50 nm particles are in the same mode; for example, on the event day May 22, GF of 50 nm particles increased from 1.08 to 1.11 and GF of 20 nm increased from 1.08 to 1.13 at the same time.

Page 3347 lines 28 and page 3348 line 12: The events require efficient enough solar radiation and low tide conditions for new particle formation. The required solar radiation makes sure that emitted iodine and organic gas compounds form secondary compounds. It is well known that secondary iodine compounds are needed for nucleation events. Because organic gas compounds are emitted from same low tide area or/and even from same seaweeds as iodine gas compounds, it is expected that also condensing secondary organic compounds are produced during the events. A part of organic compounds could be expected to form already before the events from the open ocean sources, when efficient condensation sink is missing. However, those organics would be expected to react again during the events, because of sufficient light and increased inorganic catalyst availability during the low tide events. Thus, also the condensing products originating from open ocean precursors would be formed during the events.

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Page 3349: PHA-UCPC field experiments were not carried out in the same time period as the UFO-TDMA measurements. That has been point out in the end of introduction. Overview of the instrumentation will be given in the method section and event days will be detailed in the introduction.

Minor/technical comments (referee #1)

Author comments:

We will use full words in the abstract and abbreviations will be introduced in the text of the paper.

Page 3393, line 26 will be corrected to following form: Ě., which are the products of photolysis followed by chemical reactions of biogenic iodocarbonsĚ

Page 3340: We will use word enhanced instead of word high during spring period. The concentration is enhanced compared to autumn and average values as seen from Figure 1. More detailed reference information about satellite data are only shown in acknowledgements. That information will be mentioned also in the figure caption.

Page 3340, line 14: “to exist” will be left out.

Page 3341, line 18: The required sentence will be reformulated.

Figure 10 has been described more detailed in the figure caption: HYSPLIT backward trajectories arriving to the Mace Head site at 9:00 (yellow), 11:00 (purple), 13:00 (turquoise), 15:00 (green), 17:00 (blue), and 19:00 (red) UTC (i.e. coordinated universal time), respectively, during 27 May 2002.

Figures 7-9: The time of the day (hours) will be written on the x-axis. Growth factors for 10 and 30 nm particles will be shown in the same figure.

Page 3347-3348 a reference will be added (p. 3348 line 3): Those properties (see e.g. McFiggans et al., The effect of physical and chemical aerosol properties on warm cloud droplet activation, Atmos. Chem. Phys., 6, 2593-2649, 2006) can have remarkable

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effects on the formation and properties of cloud droplets and clouds.

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