Response to Anonymous Referee #1

We would like to thank the reviewer for his/her comments. We have done our best to address each of the points as detailed below.

Note: All reviewer comments in *italics;* all responses by the authors in normal font.

The manuscript presents a modeling study of the effect of marine OM on cloud properties and the estimated aerosol indirect forcing. It is one of the first studies to do so, as most previous papers have not looked beyond the effect on CCN, and is as such well worth publishing. My main criticism is that based on the noisy-looking results (e.g. Fig 2) and the analysis of statistical significance (Fig. S1) it seems that the modeled time period may be too short to draw any real conclusions on the impact of marine OM on clouds. Furthermore, the authors do not put their results into a wider context and the methodology should be described in a bit more detail. These flaws can, however, be relatively easily fixed, and therefore I recommend publication after the following comments have been addressed.

#1. *As I already said above, the noisiness of the results (Figs 1c and 2) indicates that the model run may not be sufficiently long. This is further supported by Figure S1 which shows the areas where the modeled changes are statistically significant (using a rather high threshold p value of 0.1!): the model grids showing significant change are very scattered (the main exception being the Southern Ocean) and also frequently show up over the continents thousands of kilometers from the nearest oceans. In order to draw solid conclusions based on these simulations one should be able to explain why statistically significant changes take place in the areas they do, and not in others, but clearly this is impossible based on the current results. This leads me to think that much of the statistically significant change is in fact due to model noise. Specifically, the current results (Fig. S1) do not really support the statement that clouds in the North Pacific and Atlantic Oceans are sensitive to OM any more than e.g. parts of the Indian Ocean.*

I strongly recommend that the authors extend their key simulations for at least another five years (this probably isn't necessary for all the sensitivity simulations). While this will require a lot of computer time, I do think it necessary in order to draw robust conclusions. Furthermore, reanalyzing the extended simulation data set should be relatively straightforward and not take up too much time.

We appreciate the reviewer's comment and agree that model noise should be minimized. In the updated manuscript the model simulations G11 and G11-PI and Default and Default-PI have been extended to 10 years. Resulted changes for example in predicted aerosol indirect forcing (AIF) of -0.09 W m^{-2} now compared to -0.1 W m^{-2} before, low-level in-cloud droplet number concentration of 1.3 cm^{-3} now compared to 1.3 cm^{-3} before and liquid water path of 0.22 g m^{-2} now compared to 0.2 g m^{-2} before have been included in the modified manuscript. Corresponding changes were also made to the figures.

#2. *The introduction and methods sections are very short and lack in detail. I understand that the authors do not wish to repeat long sections of Part 1 of the manuscript. However, both parts should be able to stand alone in the sense that the readers must be able to assess the validity of the basic methodology and the significance of the results without referring to any other paper for information. Because of this, the introduction section should be expanded to summarize the main findings of previous studies on this topic and to highlight what is the added value of this*

particular study. Furthermore, a short review of the CAM5 model version used as well as the simulations performed in Part 1 (and used here in Part2) should be given.

The introduction section has been expanded and the following discussion was added to the updated manuscript: "Marine organic aerosols, emitted into the atmosphere as primary particles via bursting of bubbles at the ocean surface and secondary particles via oxidation of volatile organic compounds (VOC) such as isoprene, monoterpenes, amines, and dimethyl sulfide (DMS), have been shown to affect the chemistry and number distribution of aerosols in the marine environment (O'Dowd et al., 2004; Meskhidze and Nenes, 2006; Yoon et al., 2007; Facchini et al., 2008). Part 1 of this study (Meskhidze et al., 2011) described the implementation of marine organic aerosols in the Community Atmosphere Model version 5 (CAM5) with a 7-mode Modal Aerosol Module (MAM-7) (Liu et al., 2011). Meskhidze et al. (2011) showed that addition of marine organics led to improved agreement of the model predicted and measured concentrations of organic aerosols in the marine boundary layer, with annual average submicron aerosol mass concentration increased by up to 400 ng m^{-3} over biologically active oceanic regions. Comparison with long-term observations showed that of the two marine primary organic emission parameterizations implemented into CAM5 (Vignati et al., 2010; Gantt et al., 2011), the Gantt et al. (2011) emissions were slightly better in replicating the seasonal cycle of water insoluble organic aerosol mass concentrations. In the areas with the highest emission rates of marine organic aerosols, the cloud condensation nuclei (CCN) concentrations increased by up to 20% due to an increase in the accumulation mode (80-300 nm in diameter in CAM5) aerosol number concentration.

The potential influence of marine organic aerosols on cloud microphysical properties and radiative forcing was first discussed by Novakov and Penner (1993) and Novakov et al. (1997), who found that organic aerosols of marine origin contributed to a major fraction of marine boundary layer CCN concentration in the Tropical Atlantic. O'Dowd et al. (2004) described an increase (15-100%) in cloud droplet number concentration (CDNC) at Mace Head with the inclusion of marine organic aerosols derived from size-resolved chemistry and number distribution measurements. Ovadnevaite et al. (2011) reported similar results in an AMS study at Mace Head, finding that periods with high organic fractions and low growth factors also had a higher weighted average particle size, CCN activation efficiency, and estimated CDNC. In a modeling study, Roelofs (2008) reported results from the ECHAM5-HAM model in which marine organic aerosols increased North Atlantic CDNC by a factor of 3-4 (~35 to 120 cm^{-3}) and decreased cloud effective radius from 15-20 μm to 10-14 μm in diameter. These changes brought the model closer to satellite derived values for the region. However, not all studies examining the potential climate impact of marine organic aerosols have found that they have a large impact. Hygroscopic and CCN activity measurements of laboratory bubble bursting experiments from Fuentes et al. (2011) and Moore et al. (2011) found that despite evidence of organic compounds in sea spray aerosol, their higher hydrophobicity and lower CCN activity lead to a prediction of small changes in CCN concentration associated with marine organic aerosols and negligible impact on cloud formation. Westervelt et al. (2012) suggested that marine organic aerosols have a minor impact on climate due to GISS II-prime modeling results showing a decrease in CCN concentration (due to a decrease in particle solute concentration) in all simulation except when marine organic aerosols and sea-salt were treated as externally-mixed. Here in the second part of the study for climate forcing of marine organic aerosol, we focus on

the impact of the marine organic aerosols on cloud microphysical properties and shortwave radiative forcing."

The follow discussion was added to the methods section of the updated manuscript: "When the marine POA emissions were externally-mixed, they resulted in additional aerosol number and mass in the four model modes (Aitken, accumulation, and fine/coarse sea-salt modes) as opposed to only additional aerosol mass for the internally-mixed marine POA emissions.

The emissions from the G11 simulation in Part 1 are used in the sensitivity simulations for this work because the resulting surface concentrations had a seasonal cycle that was more similar to observations. These sensitivity simulations are carried out to determine the impact of marine organic aerosols on cloud microphysics and radiative forcing for different aerosol activation parameterizations and marine POA hygroscopicity and mixing state. Table 1 gives a summary of the 10- and 5-year long simulations with 3-month spin-up, including those performed in Part 1 on which the simulations in this work are based. Detailed descriptions of the different aerosol activation schemes, marine POA hygroscopicity and mixing state treatments, and preindustrial and present day anthropogenic emissions can be found in Meskhidze et al. (2011). In addition to simulations using the Abdul-Razzak and Ghan (2000) (hereinafter referred to as AR-G) aerosol activation parameterization from Part 1, the aerosol activation parameterization described by Fountoukis and Nenes (2005) (hereafter referred to as FN) is implemented in simulations with and without the G11 marine organic emissions ("Default-FN" and "G11-FN", respectively). An additional sensitivity test was conducted for the hygroscopicity parameter (κ) (Petters and Kreidenweis, 2007) of marine POA by increasing it (in the G11 simulation) from the $\kappa = 10^{-10}$ value used for terrestrial POA to $\kappa = 0.1$ (Liu et al., 2011). This adjusted κ represents the upper end of the potential marine POA hygroscopicity based on the measurements of κ values of 0.006 and 0.04 for estuarine (Moore et al., 2008) and riverine (Svenningsson et al., 2006) organic matter, respectively. In order to estimate the effect of marine organic aerosols on cloud radiative forcing, both the Default and G11 simulations were performed with present-day (PD) and pre-industrial (PI) aerosol and precursor emissions. The anthropogenic aerosol indirect forcing (AIF) is then calculated as the difference in model-predicted short wave cloud forcing (SWCF) between PD and PI conditions. The model simulations with PD and PI emissions used anthropogenic emissions from the IPCC AR5 dataset for the year 2000 and 1850, respectively (Bond et al., 2007; Junker and Liousse, 2008; Lamarque et al., 2010)."

#3. *The authors mention briefly that CDNC and other cloud properties show similar seasonality to Chl-a at some high-latitude areas. Given that the seasonal variation of Chl-a tends to be very large in these latitudes, it would be very interesting to see an additional figure on the seasonal changes in either CDNC or radiative forcing.*

The seasonal change in SWCF due to marine organic aerosols has been added to the updated manuscript as Fig. S2.

#4. *Overall, the introduction of the specific simulations in Section 2 could be made a bit easier to follow (e.g. give simulation name in text when specific sensitivity tests are discussed). The same goes with Table 1 which is now split into two parts (1a and 1b). In reality, many readers scan the tables and figures before deciding whether to read the text at all, and in such cases the current*

legends are confusing (not explaining why the simulations in the Part 1 paper are presented here). Either combine tables 1a and 1b or modify their legends (e.g. 1a: "— simulations run for the Part 1 paper and further analyzed here in the Part 2 paper" and 1b: "— additional CAM5 simulations —"). Indicate in Table 1a also the default activation parameterization against which FN parameterization is tested.

This has been clarified in the updated manuscript through modification of the table legends and addition of a footnote describing which aerosol activation parameterization was used.

#5. *Section 3.1.1: While it is clear based on Table 2 that LWP is not kept fixed in these simulations, reading through section 3.1.1 one might be left with this impression (see first sentences of this section). Consider reformulating.*

Discussion of constant LWP has been clarified. The updated manuscript now reads "In general it is expected that the addition of CCN may result in an increase in the in-cloud CDNC if the liquid water path (LWP) is held constant (Twomey, 1972, Albrect, 1989). Current simulations show that, when LWP is unconstrained, addition of marine organic aerosols can change both CDNC and LWP of the clouds. The effect is expected to be particularly pronounced for the low-level maritime clouds as marine aerosols are typically found within 1 km above the ocean surface (Kiliyanpilakkil and Meskhidze, 2011)."

#6. *Section 3.2.1, comparison to Meskhidze and Nenes (2006): It is speculated here that the difference in magnitude of SWCF is likely due to model grid, selected period and averaging. Given that the only differences between the simulations compared in Fig 2 (Default and G11) are marine POA, SOA and MS-, I would expect DMS to play a role as well.*

Discussion of potential influence of DMS on this comparison has been included. The updated manuscript now reads "The discrepancies in magnitude are likely to be associated with the coarser model grid, annual averaging (summertime SWCF changes of -5 to -10 W m^{-2} are predicted throughout the region), selection by Meskhidze and Nenes (2006) of a time period with a particularly large phytoplankton bloom, and the effect of DMS-derived sulfate aerosols. Since the same DMS emissions are used in the Default and G11 simulations, DMS-derived sulfate aerosols are likely to have a minor influence on the modeled changes in SWCF."

#7. *Section 3.2.1, p. 5, "Ghan et al (2011) showed —": Give the same accuracy for all the values, ie. the difference of -1.60 and -1.76 should be 0.16, not 0.2 (or alternatively -1.6, -1.8 and 0.2).*

This has been corrected in the updated manuscript.

#8. *The experiments using Vignati et al. (2010) parameterization are not discussed in the text and can thus be omitted.*

This has been corrected in the updated manuscript.

#9. *Give the names of the simulations compared in Fig 2 legend.*

This has been corrected in the updated manuscript. Figure caption in modified manuscript reads:" Model predicted percentage change between the G11 and Default in the 10-year average grid-mean column…"