

## Responses to Referee #2

### General comments:

This manuscript presents ambient concentrations of numerous organic compounds measured in marine aerosol particles, as well as a discussion of their possible origins and interrelations. Aside from the major homologous series of hydrocarbons, several molecular source tracers were quantified for the assessment of contributions from different emission sources, such as bioaerosol, biomass burning, and secondary organic aerosol (SOA). Primary biological aerosol in form of fungal spores and biogenic SOA from isoprene and pinene oxidation were shown to have important influence on the marine arctic aerosol burden. The findings from this study are valuable, since few measurements of speciated organic aerosol have been reported for marine and in particular arctic regions. The manuscript is well written, except for some grammar and spelling mistakes, and the interpretation of the presented data is reasonable. Therefore, I recommend publication of this manuscript in Biogeosciences after considering and incorporating the comments and suggestions presented below.

**Response:** We are grateful to the reviewer for the helpful comments and suggestions. Below are point-by-point responses with reviewer's comments in blue and authors' responses in black.

### Specific comments

1. Page 10433, lines 20-21: Was only one field blank collected during the entire cruise? As the authors may agree, that is not sufficient, especially during difficult sampling conditions, such as these on board of a ship. The resulting blank concentrations have, thus, no statistical basis. However, lab blanks were apparently used for blank corrections as well, which compensate at least partially for the lack of field blanks.

**Response:** We only collected one field blank during the MALINA cruise. The field blank filter, together with lab blanks, was analyzed by the same procedure as for real samples for quality assurance. The results showed no contamination for most of the measured organic species, except for minor contamination for phthalate esters and phthalic acids (less than 5% of real samples). All the data reported here were corrected for the field blank.

2. Page 10435, lines 9-11: When using the Sunset carbon analyzer, usually the NIOSH protocol (or modification thereof) is used rather than the IMPROVE method, involving thermo-optical reflectance (TOR) measurement. Please, clarify which method was really used (especially in terms of the optical charring correction) and include a reference for the method, as this is very important for comparisons of the resulting OC and EC data with those from other studies.

**Response:** We used a Sunset Lab carbon analyzer to OC and EC were determined following the Interagency Monitoring of Protected Visual Environments (IMPROVE)

thermal evolution protocol and assuming carbonate carbon in the sample to be negligible. The reference of Want et al. (2005) has been added in the revised manuscript. (see Page 5, Line 152).

3. Page 10440, lines 16-25: The authors mention several possible sources of fatty alcohols in general, but don't provide a discussion of the specific sources that may have influenced the marine aerosol in this study. For instance, biomass burning was not even an important emission source of the carbonaceous marine aerosol (as stated in the previous section), yet the authors mention it here as a possible source. It would, therefore, be more meaningful to discuss the most likely sources that influenced the marine aerosol on specific days, depending on the air mass history, as they do with other compound classes.

**Response:** Thank you. The following sentences of “Among the HMW species, C<sub>28</sub> was the dominant species (Table S1). The homologues <C<sub>20</sub> are abundant in soil microbes and marine biota, while the homologues >C<sub>24</sub> are abundant in terrestrial higher plant waxes and loess deposits. Biomass burning process can also emit a large amount of fatty alcohols and fatty acids into the atmosphere (Simoneit, 2002).” has been changed into “Among the HMW species, C<sub>28</sub> was the dominant species (Table S1). The homologues <C<sub>20</sub> are abundant in soil microbes and marine biota, while the homologues >C<sub>24</sub> are abundant in terrestrial higher plant waxes and loess deposits. Although biomass burning process can emit a large amount of fatty alcohols and fatty acids into the atmosphere (Simoneit, 2002), the contribution of biomass burning to fatty alcohols in the marine atmosphere during the MALINA campaign should be minor, a point mentioned earlier in Section 3.2.1.” in the revised manuscript. (see Page 10, Line 288-294).

4. Page 10441, lines 17-24: It is interesting to see the good correlation between mannitol and ergosterol, which has not been observed in previous studies. On the other hand, the poor correlation between arabitol and mannitol is surprising, as it has been found to be high in several other studies, such as Zhang et al., (2010a). The authors give a good explanation, though, i.e., the influence of diverse fungal sources, as the contributions are not local but due to long-range transport of fungal spores from terrestrial regions.

**Response:** The following sentences of “Arabitol and mannitol are both considered as specific tracers for fungi, while no strong correlation ( $R^2=0.48$ ,  $p<0.01$ ) was observed between them. This may imply a high diversity of fungal spores in marine aerosols collected in different locations, because different fungal species may contain different levels of arabitol and mannitol (Bauer et al., 2008).” has been reworded as: “Arabitol and

mannitol are both considered as specific tracers for fungi (Bauer et al., 2008; Zhang et al., 2010b), while no strong correlation ( $R^2=0.48$ ,  $p<0.01$ ) was observed between them. Different fungal species may contain different levels of arabitol and mannitol (Bauer et al., 2008). Such a weak correlation between arabitol and mannitol may imply a high diversity of fungal spores in the marine aerosols that were emitted not only from the Arctic Ocean but also from terrestrial regions through long-range atmospheric transport.” (see Page 11, Line 318-323).

5. Page 10446, lines 25-28: As the data presented in this section are not an actual source apportionment, it would be helpful if the authors added a statement that describes the calculation method (i.e., how the percentage values were obtained) and, thus, avoids confusion with typical source apportionment results, such as those obtained by CMB modeling.

**Response:** The following sentence has been added in the revised manuscript. “According to the above-mentioned categories, all the measured organic species were converted into their carbon contents to calculate the relative abundances of each category.” (see Page 16, Line 480-482).

#### **Technical corrections:**

1. Throughout the entire manuscript, the symbol for liters needs to be corrected, i.e., write "L" instead of "l". Also, the grammar and spelling need to be checked preferentially by a native English speaker and corrected throughout the manuscript.

**Response:** The symbol for liters was corrected throughout the revised manuscript. The authors have carefully checked the grammar and spelling problems, and hopefully will ask the handling editor of our manuscript to check it once more.

2. Page 10436, line 24: Please, add "e.g." before the reference, as this is only one of many possible studies which could be cited here.

**Response:** Corrected. (see Page 7, Line 188).

3. Page 10437, lines 1-5: The authors may also want to compare their OC/EC data with those from another recent study of marine aerosols conducted during two cruises in the Atlantic Ocean and Pacific Ocean (Zhang et al., 2010a).

**Response:** The EC contents in the Arctic aerosols collected during the MALINA campaign were very low. In most of the cases they were not detectable. Thus, the comparison OC/EC ratios with other studies are not available.

4. Page 10441, line 3: Change "innumeros" to "numerous".

**Response:** Corrected. (see Page 10, Line 301).

5. Page 10441, line 14: There is a typo in the author name "Burshtain" – it should be "Burshtein".

**Response:** Corrected. (see Page 11 Line 312).

## **References**

- Bauer, H., Claeys, M., Vermeylen, R., Schueller, E., Weinke, G., Berger, A. and Puxbaum, H.: Arabitol and mannitol as tracers for the quantification of airborne fungal spores, *Atmos. Environ.*, 42, 588-593, 2008.
- Simoneit, B. R. T.: Biomass burning-a review of organic tracers for smoke from incomplete combustion, *Appl. Geochem.*, 17, 129-162, 2002.
- Wang, H. B., Kawamura, K. and Shooter, D.: Carbonaceous and ionic components in wintertime atmospheric aerosols from two New Zealand cities: Implications for solid fuel combustion, *Atmos. Environ.*, 39, 5865-5875, 2005.
- Zhang, M., Chen, J. M., Wang, T., Cheng, T. T., Lin, L., Bhatia, R. S. and Hanvey, M.: Chemical characterization of aerosols over the Atlantic Ocean and the Pacific Ocean during two cruises in 2007 and 2008, *J. Geophys. Res.*, [Atmos], 115, doi:10.1029/2010JD014246, 2010a.
- Zhang, T., Engling, G., Chan, C.-Y., Zhang, Y.-N., Zhang, Z.-S., Lin, M., Sang, X.-F., Li, Y. D. and Li, Y.-S.: Contribution of fungal spores to particulate matter in a tropical rainforest, *Environ. Res. Lett.*, 5, doi:10.1088/1748-9326/1085/1082/024010, 2010b.