Responses to the reviewer's comments

MS No.: acp-2020-1016

Title: Seasonal characteristics of emission, distribution and radiative effect of marine organic aerosols over the western Pacific Ocean: an analysis combining observations with regional modeling

The authors greatly appreciate the valuable and constructive comments from the two reviewers, which have helped us improve the manuscript. We have addressed their comments carefully and revised the manuscript accordingly by taken their good suggestions into account. The detailed responses (blue font) are as follows:

Response to Referee #1

General comments:

In "Seasonal characteristics of emission, distribution and radiative effect of marine organic aerosols over the western Pacific Ocean: an analysis combining observations with regional modeling," Li et al. examined both primary (MPOA) and secondary (MSOA) marine organic aerosol in the western North Pacific Ocean using a regional chemistry/aerosol-climate model. Model simulated aerosol concentrations were validated against observations. Key MOA source regions and their seasonality in the western North Pacific were identified. MPOA was found to be much more important than MSOA, and significant indirect radiative effect was found. The study is thorough and covers interesting findings. However, the presentation is overtly lengthy and should be condensed. Clarification on a few key details of the analysis should also be addressed, particularly as the lack of these pieces of information prevents interpretation of the results. I have trouble understanding how the direct and indirect radiative effects were calculated in this study. Detailed comments are provided below.

Reply: Thanks for the valuable and constructive comments which help us improve the manuscript. We have responded to your comments in detail and revised the manuscript as your suggestions.

Major comments:

1. The paper would benefit from significant summarising and focusing. It may be better to summarise the model description (Sec. 2.1) in more concise terms (e.g. a summary list of all the processes included, and for the key processes, a sentence or so explaining how they are considered) and move the more detailed descriptions to the supplement. The same can also be said for the model validation (Sec. 3). While these are thoroughly done and should be documented, the shear length of the material in their entirety distracts from the storyline of the paper. Replacing with a concise summary in the main text and moving the details of the validation (with the many figures and tables) to the supplement would be helpful.

Reply: Thank you very much for your good suggestions. We revise the model description by reorganizing the sections and by moving some detailed descriptions (including formulas) to the supplement. We also reduce the length of section 3 by summarizing the model validation,

deleting detailed descriptions on validation for black carbon and gas precursors and moving relevant figures to the supplement (also suggested by the second reviewer).

2. Section 2.3.1: please add how the emitted number is determined alongside emitted mass. The discussion on size distribution between lines 263-272 may fit better here, but additional information should also be given with regards to how these fit in the size bins.
Reply: All aerosols species are assumed to have a log-normal distribution, the number concentration of aerosol species is calculated by mass concentration according to the following equation (Curci et al., 2015):

$$N_i = \frac{M_i}{\frac{4}{3}\pi r_i^3 \rho_i exp\left(\frac{9}{2} log^2 \sigma_i\right)}$$

Where r_i is the geometric mean radius of aerosol species i, σ_i is the geometric standard deviation, ρ_i is the density of aerosol species. The r_i and σ_i of MPOA are derived from the cruise measurements over the western Pacific Ocean. We add the sentence "The number concentration is calculated by mass concentration as the formula in Curci et al. (2015)" in section 2.2 in the revised version. MOA (includes MPOA and MSOA) with size larger than 0.1 μm is not considered because they only account for a very small fraction of sea spray aerosol according to cruise measurements in the western Pacific Ocean (Feng et al., 2017).

Reference

Curci, G., Hogrefe, C., Bianconi, R., Im, U., Balzarini, A., Baró, R., Brunner, D., Forkel, R., Giordano, L., Hirtl, M., Honzak, L., Jiménez-Guerrero, P., Knote, C., Langer, M., Makar, P. A., Pirovano, G., Pérez, J. L., San José, R., Syrakov, D., Tuccella, P., Werhahn, J., Wolke, R., Žabkar, R., Zhang, J., and Galmarini, S.: Uncertainties of simulated aerosol optical properties induced by assumptions on aerosol physical and chemical properties: An AQMEII-2 perspective, Atmos. Environ., 115, 541–552, 2015.

3. L773-775: one cannot make an estimation of the East Asian contribution based on a mixture of regional and global model results, especially since just above, it's noted that the regional model produced higher emission rates than global models for the same region. Note also in the abstract (L30) and conclusion (L1080).
Reply: Yes, we delete relevant comparison and discussion throughout the manuscript.

4. L801-803: as noted above, the potential difference due to different model and study time frame may be too large to draw any such conclusion about the ratio to global emission, even approximated. Was Arnold et al.'s West Pacific average similar to results from the current study?
Reply: Yes, same as the above question, we delete such comparison between regional and global estimates. Arnold et al. (2009) used two methods to estimate marine isoprene emission flux, (1) a "bottom-up" scheme using satellite products and phytoplankton-specific isoprene productivity data; (2) a "top-down" scheme by minimizing the mean bias between the model and isoprene observations in the marine atmosphere remote from the continents. Their "bottom-up" scheme is similar to the parameterization used in this study. In general, our

modeled surface atmospheric isoprene concentrations were close to their "bottom-up" results (they didn't present global isoprene emission distribution). In the Figure 2b of Arnold et al. (2009), the annual mean surface atmospheric isoprene concentrations were 0.3~5 pptv over remote West Pacific (about 25~50°N and 130~180°E), correspondingly, our model results are approximately 0.1~4 pptv over the similar region.

5. L933-934: how much do the noMOE and FULL simulations differ aside from the added MOA? In terms of properties relevant to DRE: are there differences in the wet and dry deposition that impacted other aerosol species? If yes, how much do these account for the diagnosed DRE? Given that DRE is stated to be calculated under all-sky conditions, does this mean potential differences in cloud cover due to rain suppression by MOA could also play a role? Please specify exactly which variables are used for calculating the DRE_MOA (L933-934), beyond stating that it's a subtraction of the two simulations. Related to this, how was all-aerosol DRE calculated (Table 10)?

Reply: We are sorry for the confusion. The direct radiative effect (DRE) is defined as the difference in net shortwave radiation flux at TOA (or at the surface) induced by aerosols (either individual aerosols or all aerosols, e.g., MPOA, MSOA, sea salt etc.) between cases with and without aerosols. The DRE in this study is derived by two calculations with and without aerosols (call two times in the radiation module at each time step, one with and one without aerosols) in one simulation. So, DRE reflects an instantaneous change in solar radiation fluxes induced by aerosols, without feedbacks from dry and wet depositions at this time step. The all-aerosol DRE is calculated using the same method, i.e., in the radiation module, at each time step, radiative fluxes are calculated twice with and without all aerosols and then DRE is derived from the difference between the two calculations, here all aerosols include anthropogenic aerosols internally mixed with each other and externally mixed with mineral dust and marine aerosols (sea salt, MPOA, MSOA).

The aerosol optical parameters (including extinction coefficient, single scattering albedo, and asymmetry factor) used to calculate DRE in the radiation module are derived from a Mie theory-based scheme developed by Ghan and Zaveri (2007), in which the aerosol optical parameters are pre-calculated by the Mie theory and then fitted by Chebyshev polynomials with a table of polynomial coefficients. The effect of water uptake is treated by the κ-Köhler parameterization, which calculates aerosol wet diameter due to hygroscopic growth. The bulk κ for internal mixture of aerosols is derived by the volume-weighted average of κ of each aerosol component, and the refractive index of internally mixed aerosols is calculated using the Maxwell-Garnett mixing rule. After obtaining the wet diameter and refractive index of the internally mixed aerosols (or a specific aerosol component), the aerosol optical properties can be derived from the Chebyshev fitting coefficients table. The advantage of this scheme is the much faster computational speed than traditional Mie calculation, with a similar level of accuracy. A more detailed description on parameters, method with formulas and procedure for calculating aerosol optical properties for DRE estimation is presented in a recent paper of ours (Li et al., 2020). To avoid repetition and for brevity, we briefly introduce the above method and cite this paper in the revised version.

DRE under all-sky conditions takes into account the cloud effect on clear-sky DRE (not the aerosol indirect effect on cloud nucleation and cloud properties), e.g., cloud layer at

different altitude relative to aerosol layer affects aerosol reflectivity and DRE, the DRE of scattering aerosol under all-sky condition is smaller than that under clear-sky condition (Liao and Seinfeld, 1998). So, the change in cloud cover due to rain suppression by MOA does not affect DRE estimation, the aerosol's effect on cloud and potential feedback effects are considered in the IRE calculation which is described in detail as below.

Reference
Li Jiawei, Han Zhiwei, Wu Yunfei, Xiong Zhe, Xia Xiangao, Li Jie, Liang Lin, Zhang Renjian: Aerosol radiative effects and feedbacks on boundary layer meteorology and PM2.5 chemical components during winter haze events over the Beijing-Tianjin-Hebei region. Atmos. Chem. Phys., 20, 8659–8690, 2020.
Liao, H., and J. H. Seinfeld: Effects of clouds on direct aerosol radiative forcing of climate, J. Geophys. Res., 103, 3781– 3788, 1998.

6. L977: how is IRE_MOA calculated? Also, the MSOA (L1044), sea salt and all aerosol (L1033-1043 and Table 10) IRE? I have trouble seeing how these can be properly determined from the same two simulations. Please specify the exact variables/equation/diagnostic procedure. Which processes and indirect radiative effects are captured by this definition?

Reply: We are sorry for missing the description on IRE_MOA calculation. As approaches in previous studies (Lohmann and Feichter, 2005; Wang and Penner, 2009; Leibensperger et al., 2012; Zhao et al., 2017), the indirect effect is defined as the difference in net shortwave radiation flux at TOA (or at the surface) induced by aerosols (either individual aerosols or all aerosols, e.g., MPOA, sea salt etc.) between cases with and without aerosols (or pre-industrial clean condition). The IRE due to individual or all aerosols are calculated through a series of simulations described as below:

$IRE_{mpoa}=(F\downarrow-F\uparrow)_{with\ mpoa}-(F\downarrow-F\uparrow)_{without\ mpoa}$
$IRE_{msoa}=(F\downarrow-F\uparrow)_{with\ msoa}-(F\downarrow-F\uparrow)_{without\ msoa}$
$IRE_{sea\ salt}=(F\downarrow-F\uparrow)_{with\ sea\ salt}-(F\downarrow-F\uparrow)_{without\ sea\ salt}$
$IRE_{all}=(F\downarrow-F\uparrow)_{with\ all\ aerosols}-(F\downarrow-F\uparrow)_{without\ all\ aerosols}$

Where $F\downarrow$ and $F\uparrow$ are the incoming and outgoing shortwave radiation fluxes, respectively. Here all aerosols include both natural and anthropogenic aerosols.

The indirect effect consists of the first and second indirect effect. The first indirect effect is derived from the difference in the net shortwave radiation fluxes between two calculations (call two times, one with cloud optical parameters, e.g., cloud optical depth under background condition of cloud droplet number concentration (N_c) and one with cloud optical parameters under N_c condition perturbed by aerosols, e.g., MPOA) at each time step in the radiation module, which reflects an instantaneous change in shortwave radiative flux due to aerosol perturbation to cloud properties (N_c, cloud effective radius and albedo etc.). As the commonly used method, the background condition (a pristine environment) is represented by prescribing a low bound of N_c of $10/cm^3$, which generally represent liquid stratiform cloud in clean marine conditions according to satellite observations and global model simulations (Bennartz, 2007; Hoose et al., 2009; Zeng et al., 2014; Zhao et al., 2017).

Details on calculation procedure for cloud properties and IRE are as follows:
While a specific aerosol component is considered or added (e.g. MPOA), N_c due to aerosol activation is diagnosed by the A-G scheme based on Köhler theory, then the cloud effective radius r_e is calculated as a function of N_c and cloud liquid water content following the approach of Martin et al. (1994), and the cloud optical properties (liquid cloud extinction optical depth, single scatter albedo, asymmetry factor etc.) are calculated by the scheme of Slingo et al. (1989), finally, shortwave radiation fluxes are calculated by the CCM3 radiation scheme (Kiehl et al., 1996), and the first indirect effect is derived from the difference in the net shortwave radiation fluxes between the two calculations (mentioned above) every time step within one simulation. The changes in the above cloud microphysical properties subsequently affect the conversion of cloud water to rainwater, which is a function of N_c (diagnosed above) and cloud liquid water content represented by the scheme of Beheng (1994) and further affect cloud properties, radiative fluxes and precipitation (namely the second indirect effect), and affect the first indirect effect in the next time step, so the second indirect effect influences the first indirect effect calculation through modifying cloud microphysical and optical properties (in part through altering precipitation and wet scavenging of aerosols and CCNs).

In summary, the indirect radiative effect (IRE) in this study represents processes through which aerosols perturb cloud microphysical and optical properties and solar radiation flux, that is an addition of aerosol components (e.g. MPOA) leads to increases in CCN and N_c, a decrease in r_e, and increases in cloud optical depth and cloud albedo (the first indirect effect), and leads to decreases in conversion rate from cloud water to rain water, increases in cloud water content, cloud amount, cloud optical depth and decreases in precipitation, which may in turn affect aqueous chemistry and scavenging of airborne aerosol loading and thus CCN and N_c. The first indirect effect results in a negative solar radiative effect at TOA due to increased cloud albedo and outgoing solar radiation, while the second indirect effect strengthens the negative effect.

References

Bennartz, R.: Global assessment of marine boundary layer cloud droplet number concentration from satellite, J. Geophys. Res.,112, D02201, doi:10.1029/2006jd007547, 2007.

Kiehl, J.T., Hack, J.J., Bonan, G.B., Boville, B.A., Briegleb, B.P., Williamson, D.L., Rasch, P.J.: Description of the NCAR Community Climate Model (CCM3), NCAR Technical Note, NCAR/TN-420+STR, p.152, 1996.

Hoose C., Kristja´nsson J. E., Iversen T., A. Kirkevag, Ø. Seland, and Gettelman A.: Constraining cloud droplet number concentration in GCMs suppresses the aerosol indirect effect, Geophys. Res. Lett., 36, L12807, doi:10.1029/2009GL038568, 2009.

Leibensperger E. M., Mickley L. J., Jacob D. J., Chen W.-T., Seinfeld J. H., Nenes A., Adams P. J., Streets D. G., Kumar N., and Rind D.: Climatic effects of 1950–2050 changes in US anthropogenic aerosols – Part 1: Aerosol trends and radiative forcing, Atmos. Chem. Phys., 12, 3333–3348, 2012.

Lohmann, U. and Feichter J.: Global indirect aerosol effects: a review, Atmos. Chem. Phys., 5,

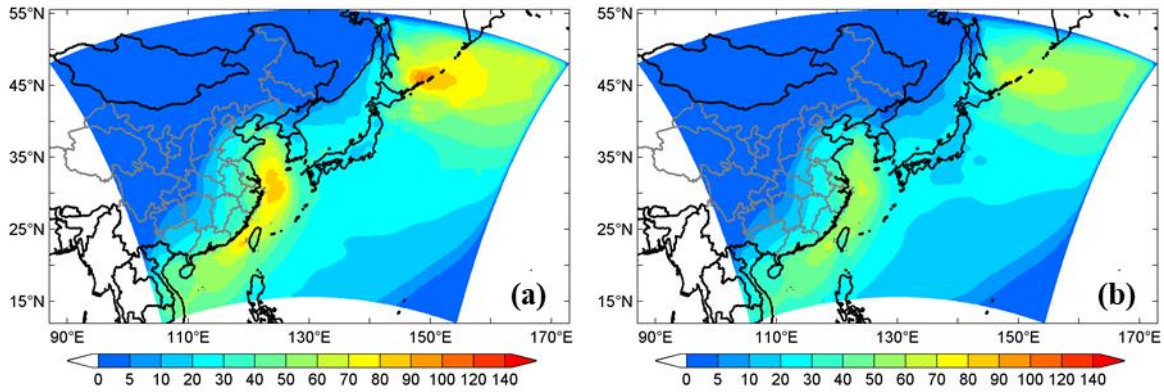
715– 737, 2005.
Martin, G. M., Johnson, D. W., and Spice, A.: The Measurements and Parameterization of Effective Radius of Droplets in Warm Stratocumulus Clouds, J. Atmos. Sci., 51, 1823–1842, 1994.
Slingo A.: A GCM Parameterization for the Shortwave Radiative Properties of Water Clouds, J. Atmos. Sci., 46(10), 1419-1427, 1989.
Wang, M., and Penner J. E.: Aerosol indirect forcing in a global model with particle nucleation, Atmos. Chem. Phys., 9, 239– 260, 2009.
Zeng, S., Riedi, J., Trepte, C. R., Winker, D. M. & Hu, Y. X.: Study of global cloud droplet number concentration with A-Train satellites. Atmos. Chem. Phys. 14, 7125–7134, 2014.
Zhao, B., Liou, K. N., Gu, Y., Li, Q., Jiang, J. H., Su, H., He, C., Tseng, H. R., Wang, S., Liu, R., Qi, L., Lee, W. L., and Hao, J.: Enhanced PM2:5 pollution in China due to aerosol-cloud interactions, Sci. Rep., 7, 4453, https://doi.org/10.1038/s41598-017-04096-8, 2017.

7. Does cloud fraction differ between the two simulations due to rain suppression? Assuming that this is included in the calculated IRE_MOA, what proportion of IRE_MOA is related to changes in cloud microphysical properties (e.g. CDNC/Nc) and how much to changes in macroscopic properties (e.g. cloud cover, precipitation)? Could any of the changes in macroscopic properties be resulting from dynamical feedback? More information would be needed to properly interpret the high IRE_MOA despite relatively low MOA concentration (for instance, compared to sea salt) and to compare to other studies.

Reply: As we discussed above, the indirect effect includes some feedback effects by the second indirect effect. To distinguish the IRE_MOA between the first indirect effect related to changes in cloud microphysical properties (e.g. CDNC/N_c) and the second indirect effect related to macroscopic properties (e.g. cloud cover, precipitation), we conduct an additional sensitivity simulation from the base case by inactivating the second indirect effect (by assigning N_c to be the background 10/cm^3 in the Beheng scheme) (i.e., just considering the first indirect effect). The contrast between the base case and the sensitivity case (Figure S5a,d) shows that the first indirect effect account for majority of the total indirect effect and the second indirect effect, which involves radiative, dynamic and precipitation feedbacks reinforces the first indirect effect (a stronger negative IRE in Figure S5a), because the cloud albedo is further enhanced resulting from increased cloud water content (together with decreased precipitation) due to weakened cloud water to rain water conversion, as clearly shown in Figure S5b,e over the western Pacific. These feedback processes are highly complex and nonlinear and beyond the scope of this manuscript, so we present a brief discussion about the relative roles of the first and second indirect effect, with the sensitivity model results and figure added in the supplement in the revised version.

The main reasons for the high IRE_MOA in this study (relevant to sea salt) could be: 1.) higher number concentration of MOA than that of sea salt (see the figure below), because the geometric mean radius of MOA (0.05μm based on cruise measurement from Feng et al., 2017) is smaller than that of the fine mode sea salt (0.1-1.0 μm); 2.) we assume a slight solubility of MOA with smaller molecule weight in this study, which could result in lower critical supersaturation for aerosol activation and more CCN. To address the uncertainty in

IRE_MOA, we conduct additional simulations regarding MOA properties. We add the sensitivity simulations, discussions and comparison between our model results and other studies, e.g., Quinn et al. (2017) in the revised version (please see more details about the sensitivity simulations in response to the question 8 below and about the comparison with Quinn et al., 2017 in the response to the second reviewer).



Model simulated annual mean near surface aerosol number concentrations for (a) MOA and (b) sea salt (units: #/cm^3).

References

Feng, L.M., Shen, H.Q., Zhu, Y.J., Gao, H.W., and Yao, X.H.: Insight into Generation and Evolution of Sea-Salt Aerosols from Field Measurements in Diversified Marine and Coastal Atmospheres, Sci. Rep., 7, 41260; doi: 10.1038/srep41260, 2017.

8. Given our limited understanding of many of MOA's properties, what's the (potential) sensitivity of the results to assumptions made in the model setup?

Reply: Thanks for raising this important question which is also our interest. Yes, our current knowledge on the physical and chemical properties of MOA is still very limited, especially over the western Pacific Ocean of east Asia, although a few cruise measurements were carried out and some knowledge on MOA properties were gained. To address this uncertainty, three additional sensitivity simulations regarding MOA properties (note we focus on MPOA due to its dominant fraction in MOA as described in the manuscript) regarding particle size, solubility and molecule weight, which is crucial to aerosol activation are conducted for the entire year to provide a range of IRE due to MOA. These sensitivity experiments, discussions, relevant figures/tables are added to the section 4.4 in the revised version as follows:

"Due to our limited knowledge on MOA properties, there cloud be uncertainties in the estimated IRE_{MOA}. To address such uncertainty, three additional sensitivity simulations from the base case (results shown in Figure S6 and Table S5 in the revised version) were carried out regarding particle size, solubility and molecule weight, which are crucial to aerosol activation (note we focus on MPOA due to its dominant fraction in MOA as shown above). The first sensitivity simulation (SENS1) assumes a smaller geometric mean radius (0.03μm instead of 0.05μm in the base case) for MPOA, resulting in a weaker domain-annual mean IRE_{MOA} (-3.5Wm^{-2}) than that in the base case (-4.2 Wm^{-2}) over the oceanic region (Figure S6b,

Table S5). The second sensitivity simulation (SENS2) assigns a lower solubility (0.03) with relatively large molecule weight (146 g mol^{-1}) for MPOA (which is similar to the properties of adipic acid, Huff Hartz et al., 2006; Miyazaki et al., 2010) instead of the slight solubility (0.1) with a smaller molecule weight (90 g mol^{-1}) (which is similar to the properties of oxalic acid, Roelofs, 2008; Miyazaki et al., 2010) in the base case, in this case, the IRE_{MOA} reduces to -2.8 Wm^{-2} (Figure S6c, Table S5). The third simulation (SENS3) combines the above two cases, assuming a smaller geometric mean radius as in SENS1 together with the lower solubility and larger molecule weight as in SENS2, it produces a further reduced IRE_{MOA} of -2.2 Wm^{-2} (Figure S6d, Table S5). The above sensitivity simulations exhibit a high sensitivity of IRE_{MOA} to the MPOA properties, and IRE_{MOA} accounts for approximately 28%, 22% and 17% of the total IRE by all aerosols in the three cases, respectively (note the total IRE also changes due to the changes in IRE_{MOA} in the sensitivity simulations), in contrast to the percentage contribution of 32% in the base case".

References

Huff Hartz Kara E., Tischuk Joshua E., Chan Man Nin, Chan Chak K., Donahue Neil M., Pandis Spyros N.: Cloud condensation nuclei activation of limited solubility organic aerosol, Atmospheric Environment 40, 605–617, 2006.

Miyazaki Yuzo, Kawamura Kimitaka, and Sawano Maki: Size distributions and chemical characterization of water-soluble organic aerosols over the western North Pacific in summer, J. Geophys. Res., 115, D23210, doi:10.1029/2010JD014439, 2010.

More minor comments:

9. A suggestion for consideration, since from my understanding, ACP does not limit the abstract length: the abstract as it stands right now reads more like a conclusion/summary. Could it be pared down more? (for instance, what are the three key findings of this study?)

Reply: Thank you for the suggestion. We revise the abstract by summarizing and highlighting main findings from this study.

10. The title states "…an analysis combining observations with regional modeling," but as I understand it, the observations were not used to bias correct or calibrate the model in any way. In this sense the observations were used purely for model validation, and as such this wording is perhaps misleading (there's no "combining" involved). Instead, perhaps something more along the lines of "model validation and regional modeling" would be more accurate, if the authors decide to retain a focus on the model validation part.

Reply: Yes, the observations are mainly for model validation and interpretation, we revise the title to "Seasonal characteristics of emission, distribution and radiative effect of marine organic aerosols over the western Pacific Ocean: an investigation with a coupled regional climate-aerosol model".

11. L145-147: "supposed to" is not a good word choice here. If one wants to express uncertainty, "may" could be a good replacement. Reference(s) for this claim should also be added.

Reply: revised.

12. L166-168: the internally mixed anthropogenic aerosol is assumed to have a fixed distribution that does not change shape following activation and sedimentation? If yes, has there been studies justifying this choice? I wonder if the anthropogenic aerosol may be represented by size bins, as is done for the natural aerosols. If yes, please clarify.

Reply: In the current version of RIEMS-Chem, a bulk method is used to represent internally mixed anthropogenic aerosols, with a typical geometric diameter (standard deviation) fitted by a lognormal distribution based on recent observations in east China (Ma et al., 2017; Wu et al., 2017), which found that the geometric mean radius of a dry aerosol internal mixture during the periods from light/moderate to severe pollution stages increased slightly from 0.10 to 0.12 μm, so a geometric mean radius of 0.11 μm with a standard deviation of 1.65 are chosen for the internal mixture of anthropogenic aerosols, as that in Li et al. (2020).

References

Ma, Q. X., Wu, Y. F., Zhang, D. Z., Wang, X. J., Xia, Y. J., Liu, X. Y., Tian, P., Han, Z. W., Xia, X. A., Wang, Y., and Zhang, R. J.: Roles of regional transport and heterogeneous reactions in the PM2.5 increase during winter haze episodes in Beijing, Sci. Total. Environ., 599/600, 246–253, 2017.

Wu, Y. F., Wang, X. J., Tao, J., Huang, R. J., Tian, P., Cao, J. J., Zhang, L. M., Ho, K. F., Han, Z. W., Zhang, R. J.: Size distribution and source of black carbon aerosol in urban Beijing during winter haze episodes, Atmos. Chem. Phys., 17, 7965-7975, 2017.

Li Jiawei, Han Zhiwei, Wu Yunfei, Xiong Zhe, Xia Xiangao, Li Jie, Liang Lin, Zhang Renjian: Aerosol radiative effects and feedbacks on boundary layer meteorology and PM2.5 chemical components during winter haze events over the Beijing-Tianjin-Hebei region. Atmos. Chem. Phys., 20, 8659–8690, 2020.

13. L168: “Natural aerosols (mineral dust and sea salt)” and MPOA? Also, in general, how do MPOA and MSOA fit in these model descriptions? E.g. on L175: what’s the hygroscopicity of MPOA and MSOA? The same as POA and SOA?

Reply: We revise the sentence to “Mineral dust and sea salt are represented by ……”. The hygroscopicity for MPOA (0.1) and MSOA (0.2) are assumed to be the same as those for anthropogenic POA and SOA, we add relevant information in section 2.2 in the revised version.

14. L210: OMss is the organic mass fraction of sea spray aerosol, not sea salt.

Reply: Yes, revised.

15. L220-221: please add reference for the OM/OC ratio

Reply: Added.

Reference

Gantt, B. and Meskhidze, N.: The physical and chemical characteristics of marine primary organic aerosol: a review, Atmos. Chem. Phys., 13, 3979–3996, 2013.

16. L262: please add reference/justification of choice for the MSOA soluble mass fraction

Reply: So far, our knowledge on the properties of marine organic aerosols is very limited, especially in the western Pacific Ocean. Facchini et al. (2008) found that the OC within submicron particles generated by bubble bursting is mainly water insoluble (on average 94 ± 4% of total carbon) based on bubble bursting experiments during a phytoplankton bloom in the North Atlantic, which also denotes approximately 10% of OC is soluble.

In this study, we assume that MPOA is slightly soluble with a solubility of 0.1 considering aging processes it may undergo (Gantt and Meskhidze, 2013). Because SOA is more hygroscopic than POA, with respect to that the hygroscopicity (κ) of SOA is about twice that of POA (Liu and Wang, 2010; Westervelt et al., 2012), we assume the solubility of MSOA is twice that of MPOA. The uncertainty in chemical properties of MOA and its potential effect on IRE is discussed above and in the response to the second reviewer.

References

Facchini, M. C., Rinaldi, M., Decesari, S., Carbone, C., Finessi, E., Mircea, M., Fuzzi, S., Ceburnis, D., Flanagan, R., Nilsson, D., de Leeuw, G., Martino, M., Woeltjen, J., and O'Dowd, C. D.: Primary sub-micron marine aerosol dominated by insoluble organic colloids and aggregates, Geophys. Res. Lett., 35, L17814, doi:10.1029/2008GL034210, 2008.

Liu X. and Wang J.: How important is organic aerosol hygroscopicity to aerosol indirect forcing, Environ. Res. Lett. 5, 044010, doi:10.1088/1748-9326/5/4/044010, 2010.

Westervelt, D. M., Moore, R. H., Nenes, A., and Adams, P. J.: Effect of primary organic sea spray emissions on cloud condensation nuclei concentrations, Atmos. Chem. Phys., 12, 89–101, 2012.

17. L282-283: perhaps I'm missing something, but if aerosol activation/Nc is already calculated based on the Abdul-Razzak and Ghan scheme, why is "the number of aerosols activated assumed to be equal to the number of aerosols scavenged in cloud"?

Reply: We are sorry for the confusion. What we want to express is 'the activated aerosols (into cloud droplet) are removed from the air, the number of removed airborne aerosol is equal to that activated", we revised the relevant sentence.

18. L359: Pearson correlation coefficient? Please be specific.

Reply: Yes.

19. L718-719 and Figure 7: how does the sea salt emission look like? Perhaps add a column in figure 7 for sea salt emission (since EM_POA= α×Ess×OMss)?

Reply: Thank you, we add a column in Figure 7 for sea salt emission following your suggestion in the revised version.

20. L756-759: please clarify if this is a speculation or confirmed by analysis. It's difficult to tell by the wording.

Reply: We check the data and find that in the NWP region, the Chl-a concentration is negatively correlated with sea salt emission in MAM, whereas they are positively correlated

in SON. This could lead to the slightly higher MPOA emission (related to both Chl-a concentration and sea salt emission) in SON than that in MAM. We have revised the relevant description in the manuscript.

21. L777: “annual mean” as in "ng m-2 s-1" averaged over the area and over the whole year? Or “annual emission” in "Tg y-1"?
Reply: It is “annual emission in Tg y^{-1}”.

22. L788-790: what is the relevance of this sentence in the context of the current study? Does this then imply that it is reasonable to compare simulation to observation from a different year for order of magnitude check?
Reply: This sentence is not relevant to the model results, so we delete it in the revised version.

23. Figures 4 and 5: do the standard deviations represent variability of the monthly/seasonal mean across multiple years or also the variability within each month/season?
Reply: We draw the standard deviations in Figures 4 and 5 based on raw data presented in the publication, these data were sampled on a bi-weekly basis in the islands, so the deviation could represent bi-weekly variability.

24. Figure 9: “mean monthly” (cm month-1; total monthly precipitation averaged over multiple months) instead of “monthly mean” (mm h-1; average precipitation rate over each month). Note both in figure title and caption.
Reply: Yes, it is an average over three months in each season, we revise it both in figure title and caption in the revised version.

25. Table 9: “cm grid-1 month-1”: do the authors mean cm month-1?
Reply: It means “the sum of gridded monthly total precipitation divided by the total grid number in a specific region, so it represents cm $month^{-1}$ for a certain region”. The unit is revised.

Response to Referee #2

General comments:
This study focuses on primary and secondary marine aerosols over the Western Pacific Ocean and their radiative effect. The findings are interesting and this study is well suited for ACP. In its present form, I found this study is too lengthy which makes it challenging to read. Important methodological information and analysis are missing regarding the radiative impact of MOA.
Reply: Thank you for your pertinent and valuable comments which help us improve the manuscript. We address your comments carefully and revise the manuscript accordingly by taking your suggestions into account.

Abstract is very long. I recommend the authors try to highlight no more than 2-3 key points.
Reply: Thank you. We revise the abstract by highlighting several key points.

line 25. It reads like a new finding but as far as I understand this just describes the parameterization of Gantt et al. This should be clarified

Reply: Sorry for the description, we delete this sentence in the revised version.

line 70. The introduction focuses primarily on literature published prior to 2012. A lot has been done both in terms of observations (field and lab) and in terms of parameterization since that needs to be discussed by the authors.

Here are a couple of studies (by no mean an exhaustive list) that the authors may want to consider

Conte, L., Szopa, S., Aumont, O., Gros, V., & Bopp, L. (2020). Sources and sinks of isoprene in the global open ocean: Simulated patterns and emissions to the atmosphere. Journal of Geophysical Research: Oceans, 125, e2019JC015946. https://doi.org/10.1029/2019JC015946

Bates, T. S., Quinn, P. K., Coffman, D. J., Johnson, J. E., Upchurch, L., Saliba, G., et al. (2020). Variability in Marine Plankton Ecosystems Are Not Observed in Freshly Emitted Sea Spray Aerosol Over the North Atlantic Ocean. Geophysical Research Letters, 47, e2019GL085938. https://doi.org/10.1029/2019GL085938

Quinn, P. K., Bates, T. S., Coffman, D. J., Upchurch, L., Johnson, J. E., Moore, R., et al. (2019). Seasonal variations in western North Atlantic remote marine aerosol properties. Journal of Geophysical Research: Atmospheres, 2019; 124: 14240–14261. https://doi.org/10.1029/2019JD031740.

Brüggemann, M., Hayeck, N. & George, C. Interfacial photochemistry at the ocean surface is a global source of organic vapors and aerosols. Nat Commun 9, 2101 (2018). https://doi.org/10.1038/s41467-018-04528-7

Betram et al. Sea spray aerosol chemical composition: elemental and molecular mimics for laboratory studies of heterogeneous and multiphase reactions. Chemical Society Reviews, 31 Mar 2018, 47(7):2374-2400 DOI: 10.1039/c7cs00008a

Quinn, P., Coffman, D., Johnson, J. et al. Small fraction of marine cloud condensation nuclei made up of sea spray aerosol. Nature Geosci 10, 674–679 (2017). https://doi.org/10.1038/ngeo3003

S. M. Burrows and O. Ogunro and A. A. Frossard and L. M. Russell and P. J. Rasch and S. M. Elliott A physically based framework for modeling the organic fractionation of sea spray aerosol from bubble film Langmuir equilibria Atmospheric Chemistry and Physics 14, 13601–13629 (2014). https://doi.org/10.5194/acp-14-13601-2014

Reply: We are sorry for missing these papers, and thank you very much for telling us recent progresses on this issue in both observation and parameterization. They are valuable and helpful for this study.

We revise the introduction in the revised version by including these references in the revised version, which is described as “However, Quinn et al. (2014) found that the organic carbon content of sea spray aerosol is weakly correlated with satellite retrieved chlorophyll-a concentration based on cruise measurements in the North Atlantic Ocean and the coastal waters of California. Bates et al (2020) reported that plankton bloom has little effect on the emission flux, organic fraction or cloud condensation nuclei of sea spray aerosol based on cruise experiment over the North Atlantic. Burrows et al. (2014) developed a novel physically

based framework for parameterizing the organic fractionation of sea spray aerosol by consideration of ocean biogeochemistry processes, and their predicted relationships between Chl-a and organic fraction are similar to existing empirical parameterizations associated with ocean Chl-a concentrations at high Chl-a levels, but the empirical relationships may not be adequate to predict OM fraction of sea spray aerosol outside of strong seasonal blooms. Considering the strong bloom seasonality in the western Pacific region and the availability of global satellite data for Chl-a concentration, and the lack of cruise measurements on the relationship between sea spray organic aerosol fluxes and Chl-a in this region, we adopted the scheme of Gantt et al (2011) for parameterizing marine primary organic aerosol emission in this study."

We also add publications on CCN activity of sea spray aerosol as "Based on the measurements from seven research cruises over the Pacific, Southern, Arctic and Atlantic oceans between 1993 and 2015, Quinn et al. (2017) indicated that sea spray aerosol generally makes a contribution of less than 30% to CCN population at supersaturation of 0.1 to 1.0% on a global basis"

The revised introduction includes more relevant studies on this issue, although their findings or conclusions may be different.

line 169 It would be worth mentioning that this range exceeds the valid range for Monahan ($0.8 < r80 < 10$mm)

Reply: We are sorry for missing the relevant information. We actually apply the scheme of Gong (2003) in this study, which was an improvement of the Monahan et al. (1986) scheme. The valid range in the Gong (2003) scheme is from 0.07μm to 20μm radius at RH = 80%. We add the description and relevant reference in the revision.

Reference

Gong, S.L., 2003. A parameterization of sea-salt aerosol source function for sub- and super-micron particles, Global Biogeochem. Cy., 17(4), 1097. doi:10.1029/2003GB002079.

line 202 GEIA is a portal for many different inventories. Are the authors using a climatology of MEGANv2? That would be surprising since very detailed year-specific inventories are used for anthropogenic and biomass burning emissions. Please clarify.

Reply: Sorry for the confusion. The biogenic VOC emission is derived from the CAMS-BIO Global biogenic emissions dataset (CAMS-GLOB-BIO v3.1) (Granier et al., 2019; Sindelarova et al., 2014) distributed by ECCAD-GEIA (https://permalink.aeris-data.fr/CAMS-GLOB-BIO, last access: 2020/02/10) and the monthly mean biogenic emission for the year 2014 with a horizontal resolution of 0.25° is used, which is consistent with other year-specific inventories. We revise relevant description and add the references in the revision.

References

Granier, C., S. Darras, H. Denier van der Gon, J. Doubalova, N. Elguindi, B. Galle, M. Gauss, M. Guevara, J.-P. Jalkanen, J. Kuenen, C. Liousse, B. Quack, D. Simpson, K. Sindelarova, 2019. The Copernicus Atmosphere Monitoring Service global and regional emissions (April

2019 version), Report April 2019 version, doi:10.24380/d0bn-kx16.
Sindelarova, K., Granier, C., Bouarar, I., Guenther, A., Tilmes, S., Stavrakou, T., Müller, J.-F., Kuhn, U., Stefani, P., and Knorr, W., 2014. Global data set of biogenic VOC emissions calculated by the MEGAN model over the last 30 years. Atmos. Chem. Phys., 14, 9317–9341. https://doi.org/10.5194/acp-14-9317-2014.

line 226 It seems there could be a lot of other reasons for this difference. MODIS vs VIIRS Chl-A, differences in wind speed and sea salt parameterizations.
Reply: Yes, we delete "The large difference in the choice of α suggests that the emission rate of MPOA could be very regionally dependent" and move the description of the scheme to the support material in the revised version.

line 229 This section completely ignores the abiotic source of isoprene (see references above), which may be as large if not larger than the biological source in marine environments.
Reply: Thank you. We only consider marine isoprene from biological source in this study. The marine abiotic source of isoprene (due to photochemical production in the sea surface microlayer) may be important according to recent studies (Brüggemann et al. 2018; Conte et al., 2020), however, the production mechanism is still highly uncertain. Brüggemann et al. (2018) estimated a global total oceanic isoprene emission of 1.11 Tg yr^{-1} from both biological and photochemical production. It appears that the scheme of Gantt et al. (2009) predicted a higher biological isoprene emission, because their estimate (0.92 Tg yr-1) is close to that in Brüggemann et al. (2018). We add a description on the abiotic source of isoprene with relevant publications in section 2.3 in the revised version as "The marine abiotic source of isoprene (due to photochemical production in the sea surface microlayer) may be important according to recent studies (Brüggemann et al., 2018; Conte et al., 2020), which is not considered in this study because the production mechanism for marine abiotic isoprene is poorly understood at present."

line 265 I may be missing something but I am not sure how to reconcile the source function for sea salt (radius > 0.1um) which is used to derive MOA emissions, with the assumed diameter of MOA (0.1 um). Please clarify.
Reply: we are sorry for missing such information. As we explained above, we use the scheme of Gong (2003) developed based on Monahan et al. (1986), so the sea salt radius range is from 0.07μm to 20μm. Only fine model MOA (~0.1μm) is considered in this study.

line 267 do the authors also use 5 size bins to represent MPOA or do they only consider sub-micron MPOA for this work?
Reply: We only consider the sub-micron MPOA because the cruise measurements over the marginal seas of China and the western Pacific (Feng et al., 2017) revealed that TOC mass (mainly in MPOA) is mainly concentrated in the sub-micron size and the super-micron TOC was generally below the detection limit.

Section 3. I suggest to have a section devoted solely to describing the different sources of observations, such that section 3 can focus solely on the model performance. The analysis of

BC should focus more clearly on how these observations can help understand marine organic aerosols. The detailed BC analysis presented here could be moved to supporting materials, which would help shorten the manuscript.

Reply: Thank you for the good suggestion. We add section 2.5 for describing the different sources of observations, and shorten the manuscript by moving Figure 4 (in previous version) to the supplement and by deleting relevant description on model validation for BC.

It would be helpful to evaluate the simulated Na+ with the UMiami/Prospero dataset.

Reply: Yes, following your suggestion, we collected bi-weekly Na+ measurements at Japan islands from the Acid Deposition Monitoring Network in East Asia (EANET) and compared with our model simulations. We add a brief description on this comparison in section 3.1 in the revision as "Comparison with observations of Sodium (Na+) concentration at 6 Japan coastal/island sites from EANET is conducted to further examine the model performance for sea salt. The modeled sodium is estimated to be 38.56% of sea salt mass (Kelly et al., 2010), and the agreement between observation and model simulation is generally satisfactory at all sites except at Oki in December, when the model largely underpredict Na+. The model well reproduces the seasonality of sodium concentration, with the maximum in winter and the minimum in summer (Figure 3). The model predicts sodium concentration best at Ogasawara, with the correlation coefficient of 0.85 and NMB of 5%. The overall correlation coefficient for all sites is 0.50, with NMB of -11% (Table S1)". Figure 3 (below) and a table for statistics in the supplement (Table S1) are also added in the revised version.

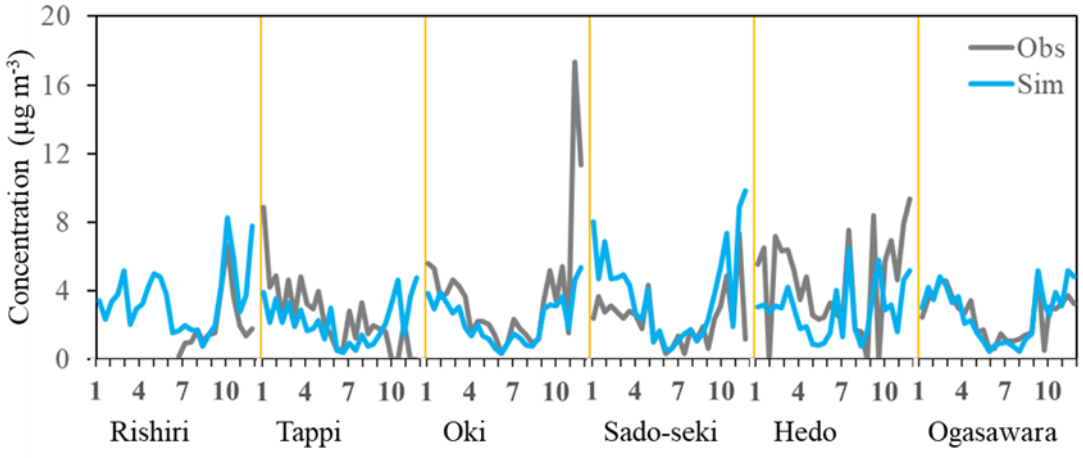


Figure 3 Observed and model simulated sodium (Na+) concentrations at 6 coastal/island EANET sites in Japan for the year 2014. The x axis is month for each site.

Reference

Kelly J. T., Bhave P. V., Nolte C. G., Shankar U. and Foley K. M.: Simulating emission and chemical evolution of coarse sea-salt particles in the Community Multiscale Air Quality (CMAQ) model. Geosci. Model Dev., 3, 257–273, 2010.

line 385-396. Suggests removing or moving to supporting materials

Reply: Yes, we move this part to supporting materials (Table S2 in the revision).

line 471 The model does not seem to capture the variability in OA from 4/7 to 4/13 (e.g., it shows high values in 4/11 for instance). Could the authors comment on this discrepancy? Could the model underestimate land SOA (which will not correlate with BC) over this time period?
Reply: Yes, the model overpredicts OA on 11 and 13 April. We check model results carefully and find the main reason for the overprediction is the model bias in prediction of wind direction, e.g, the observed wind direction is northeast wind on 13 April, whereas the model simulation is north wind, which bring in land OA from Japan and marine OA from bloom regions north of the ship, leading to overprediction of OA concentration. Wind prediction on 10 and 12 April (when the two OC peaks occur) is consistent with observation. The modeled land SOA is much smaller than land POA, but we haven't SOA observation to compare with model results.

line 478 This needs some reference. What is the size range of fungi spores?
Reply: The size range of fungal spores is ~5μm for different sources (Fröhlich-Nowoisky et al. 2016).

References
Janine Fröhlich-Nowoisky, Christopher J. Kampf, Bettina Weber, J. Alex Huffman, Christopher Pöhlker, Meinrat O. Andreae, Naama Lang-Yona, Susannah M. Burrows, Sachin S. Gunthe, Wolfgang Elbert, Hang Su, Peter Hoor, Eckhard Thines, Thorsten Hoffmann, Viviane R. Després, Ulrich Pöschl: Bioaerosols in the Earth system: Climate, health, and ecosystem interactions. Atmospheric Research, 182, 346-376, 2016.

line 583. Many aspects of the overall OA budget remain challenging to represent (https://acp.copernicus.org/articles/20/2637/2020/). The contributions of MOA is fairly small at most sites. Could optimization of the land source of SOA or the removal of OA also reduce the model bias?
Reply: Because the land source of SOA is generally underpredicted by current CTM models due to potential uncertainties in SOA sources, chemical mechanisms and removal parameterizations, the optimization of the land source of SOA may also reduce the model biases, in addition to considering marine MOA. We revise the relevant sentence to "It was striking that the inclusion of marine-OC obviously improved the model performance, reducing the NMB from -21% to -3%, although the improvement of prediction for SOA from land source may also reduce the model bias at the Huaniao island".

line 626. Does MOZART include MOA emissions?
Reply: MOZART-4 does not include MOA emissions.

line 711. I suggest to also compare with satellite AOD (MODIS, MISR, VIIRS) so that performances over the Western Pacific Ocean can be better assessed.
Reply: Thank you for the suggestion. We collected VIIRS satellite retrievals and compared with the modeled AOD at 550 nm, the model is generally capable of reproducing AOD

distribution and magnitude in the study domain, but AOD reflects the column integrated extinction coefficient due to all aerosols, we add relevant discussion in the text and put the comparison figure in the supplement (Figure S2) in the revision.

lines 771-775. You cannot mix your regional estimate with previous global estimate. Instead you would need to run your model globally to draw such conclusion. This also means that the abstract and conclusion need to be revised. Same issue on line 801.
Reply: Yes, we delete all the relevant comparison and description in the revised version.

line 885. Please clarify why this is impressive.
Reply: "It was impressive" to "It was found".

Sections 4.3 and 4.4
While there is an excess of details in previous sections, more analysis/method descriptions are needed here.
Reply: Thank you for the suggestion. We add detailed descriptions on the methods for calculating DRE and IRE and more analysis in section 4.3 and 4.4 in the revised version.

Please provide the equations to estimate DRE and IDRE. It seems that you would need more than 2 experiments to estimate the IDRE for the different types of aerosols.
Reply: We are sorry for the confusion and mistakes. We present a detailed description on how to calculate the DRE and IRE in our responses (please see our response to the first reviewer who raise the same question) and add these descriptions in section 4.3 and 4.4 in the revised version.

Are these estimates based on an ensemble of 1 yr simulations? Are the differences shown here significant (relative to natural variability)? In general the authors need to better quantify the uncertainties associated with their estimates? This is especially important for the IRE_MOA. The authors also need to discuss their findings in the context of recent work that suggests a small role of SSA for CCNs (e.g., Quinn et al. DOI: 10.1038/NGEO3003)
Reply: Yes, we conduct a 1 yr simulation for MOA in this study because most of the observations, including the cruise measurements are available for the year 2014 and the limitation in computational resources for the on-line coupled model simulation, we plan to conduct multi-years simulation to explore inter-annual variation relative to natural variability in the future.
We realize that the model results could be subject to uncertainties due to our limited knowledge on the properties of MOA, to explore and quantify this uncertainty, we conducted additional sensitivity simulations with respect to the size, solubility and molecule weight and present a range of IRE_MOA depending on MOA properties. These experiments, discussions, relevant figures/tables are added to the section 4.4 in the revised version as follows:
"Due to our limited knowledge on MOA properties, there cloud be uncertainties in the estimated IRE_{MOA}. To address such uncertainty, three additional sensitivity simulations from the base case (results shown in Figure S6 and Table S5 in the revised version) were carried out regarding particle size, solubility and molecule weight, which are crucial to aerosol

activation (note we focus on MPOA due to its dominant fraction in MOA as shown above). The first sensitivity simulation (SENS1) assumes a smaller geometric mean radius (0.03μm instead of 0.05μm in the base case) for MPOA, resulting in a weaker domain-annual mean IRE_{MOA} (-3.5Wm^{-2}) than that in the base case (-4.2 Wm^{-2}) over the oceanic region (Figure S6b, Table S5). The second sensitivity simulation (SENS2) assigns a lower solubility (0.03) with relatively large molecule weight (146 g mol^{-1}) for MPOA (which is similar to the properties of adipic acid, Huff Hartz et al., 2006; Miyazaki et al., 2010) instead of the slight solubility (0.1) with a smaller molecule weight (90 g mol^{-1}) (which is similar to the properties of oxalic acid, Roelofs, 2008; Miyazaki et al., 2010) in the base case, in this case, the IRE_{MOA} reduces to -2.8 Wm^{-2} (Figure S6c, Table S5). The third simulation (SENS3) combines the above two cases, assuming a smaller geometric mean radius as in SENS1 together with the lower solubility and larger molecule weight as in SENS2, it produces a further reduced IRE_{MOA} of -2.2 Wm^{-2} (Figure S6d, Table S5). The above sensitivity simulations exhibit a high sensitivity of IRE_{MOA} to the MPOA properties, and IRE_{MOA} accounts for approximately 28%, 22% and 17% of the total IRE by all aerosols in the three cases, respectively (note the total IRE also changes due to the changes in IRE_{MOA} in the sensitivity simulations), in contrast to the percentage contribution of 32% in the base case".

References

Huff Hartz Kara E., Tischuk Joshua E., Chan Man Nin, Chan Chak K., Donahue Neil M., Pandis Spyros N.: Cloud condensation nuclei activation of limited solubility organic aerosol, Atmospheric Environment 40, 605–617, 2006.

Miyazaki Yuzo, Kawamura Kimitaka, and Sawano Maki: Size distributions and chemical characterization of water-soluble organic aerosols over the western North Pacific in summer, J. Geophys. Res., 115, D23210, doi:10.1029/2010JD014439, 2010.

Thank you for introducing Quinn et al. (2017) to us, "It is interesting to note that Quinn et al. (2017) indicated that sea spray aerosol generally makes a contribution of less than 30% to CCN population at supersaturation of 0.1 to 1.0% on a global basis based on measurements onboard seven research cruises over the Pacific, Southern, Arctic and Atlantic oceans. Our study appears to predict a higher contribution of sea spray aerosol (the sum of MOA and sea salt) to the total indirect effect (59% in the base case) in the western Pacific region. Several factors may lead to the differences between this study and Quinn et al. (2017). One of the major reasons could be the different MOA properties assigned for activation calculation based on Köhler theory; as discussed above, we assume MPOA to be slightly soluble, considering that MPOA may undergo aging and a part of MPOA could be hydrophilic, whereas Quinn et al. (2017) assumed marine particulate organic matter was insoluble. Our sensitivity simulations show the MPOA chemical properties considerably affect IRE estimation, while a smaller size with a lower solubility and a larger molecule weight are assigned for MPOA, the IRE_{MOA} is noticeably reduced and the contribution of sea spray aerosol to the total IRE by all aerosols become 47% (with contributions from MOA and sea salt being 17% and 30%, respectively). Another reason could be associated with the assumption of the mixing state between MPOA and other aerosols, an external mixture of MPOA and sea salt is assumed in this study, which means additional marine organic aerosols are produced to affect cloud

properties and represents an upper limit of indirect effect, whereas an internal mixing could be assumed in Quinn et al. (2017). In addition, the study domain is different in the two studies, although some cruise measurements have been carried out and a few understandings on MOA properties (e.g., size distribution) was gained, the observation and analysis for MOA chemical properties are so far almost absent in the western Pacific of East Asia. The western Pacific Ocean is just downwind of the East Asian continent, which have large amounts of anthropogenic aerosols, mineral dust, and nutrients inputs to the marginal seas of China from the Yangtze and Yellow Rivers, could be very different from remote clean oceans in the world, therefore marine biogeochemistry, marine aerosol sources and properties, as well as their potentials to be CCN and impacts on radiation, cloud, and precipitation deserve further investigation in the future".
We add the above discussions in section 4.4 in the revised version

Thanks again for the comments and suggestions

Next is the supplementary material (in the revised version)

1. Marine primary organic aerosol (MPOA) emission

The emission rate of MPOA is the product of sea spray aerosol (SSA) emission rate (E_{SSA}) and organic matter fraction of sea spray aerosol (OM_{SSA}, unitless in a range of 0~1), i.e.

$$E_{MPOA} = \alpha \times E_{SSA} \times OM_{SSA} \quad (1),$$

where OM_{SSA} is expressed as a function of wind speed, surface seawater Chl-a concentration, and aerosol size, and α is a tuning factor. The calculation of OM_{SSA} follows the method of Gantt et al. (2012):

$$OM_{SSA} = \frac{\left(\frac{1}{1+exp\,(X(-2.63[Chl-a])+X(0.18U_{10}))}\right)}{1+0.03\,exp\,(6.81D_p)} + \frac{0.03}{1+exp\,(X(-2.63[Chl-a])+X(0.18U_{10}))} \quad (2),$$

where U_{10} is wind speed at 10 meter (m s^{-1}) simulated online by RIEMS-Chem, D_p is the diameter of sea salt aerosol, and Chl-a is the surface seawater chlorophyll-a concentration (mg m^{-3}). The Level-3 daily mean Chl-a concentration retrievals with 9 km resolution from the VIIRS (Visible infrared Imaging Radiometer) sensor onboard the Suomi National Polar-orbiting Partnership (SNPP) satellite platform (OBPG, 2018) are obtained and used for model inputs and it can reflect day-to-day variation of sea surface Chl-a concentration associated with phytoplankton bloom in the western Pacific. X is a unitless adjustable coefficient and is set to 3 based on Gantt et al. (2012).

For the tuning factor α, Gantt et al. (2012) suggested a factor of 6 was able to minimize the relative model biases for the global model GEOS-Chem at two oceanic sites (Mace Head in North Atlantic and Amsterdam Island in remote south Indian Ocean). In this study, we found that a factor of 2 was optimal to obtain the least bias between model simulation and observation over the western Pacific.

2. Marine isoprene emission

The sea-air flux of marine isoprene (E_{isop} in the unit of μg m^{-2} s^{-1}) is parameterized following the method of Palmer and Shaw (2005), which can be expressed as:

$$E_{isop} = k \times SW_{isop} \quad (3),$$

where k is the sea-air exchange coefficient (cm h^{-1}) and is calculated as:

$$k=0.31\times U_{10}\times(660/Sc)^{1/2} \quad (4),$$

where Sc is the Schmit number of Isoprene.

The surface seawater isoprene concentration SW_{isop} (μg m^{-3}) related to phytoplankton activities is parameterized by the scheme of Gantt et al. (2009):

$$SW_{isop} = H_{\max} \times [Chl\text{-}a] \times \int_0^{H_{\max}} EF \ln(I)^2 dh \quad (5),$$

where EF is the emission factor of isoprene released by phytoplankton, I is the ambient photosynthetically active radiation (PAR, in the unit of μEm^{-2} s^{-1}), H_{max} is the total water depth which isoprene production can occur from the surface to the point and is calculated as:

$$H_{\max} = -\ln(\frac{2.5}{I_0})\frac{1}{k_{490}} \quad (6),$$

where I_0 is the all-sky surface incoming solar radiation (W m^{-2}) provided by the model during simulation. I_0 and I have an approximate relationship of 1 W m^{-2} ≈ 2 μEm^{-2} s^{-1}. The diffuse attenuation coefficient values at 490 nm k_{490} (m^{-1}) is also obtained from VIIRS satellite. The isoprene production is assumed to occur when the light level is greater than 2.5 W m^{-2} in surface sea water.

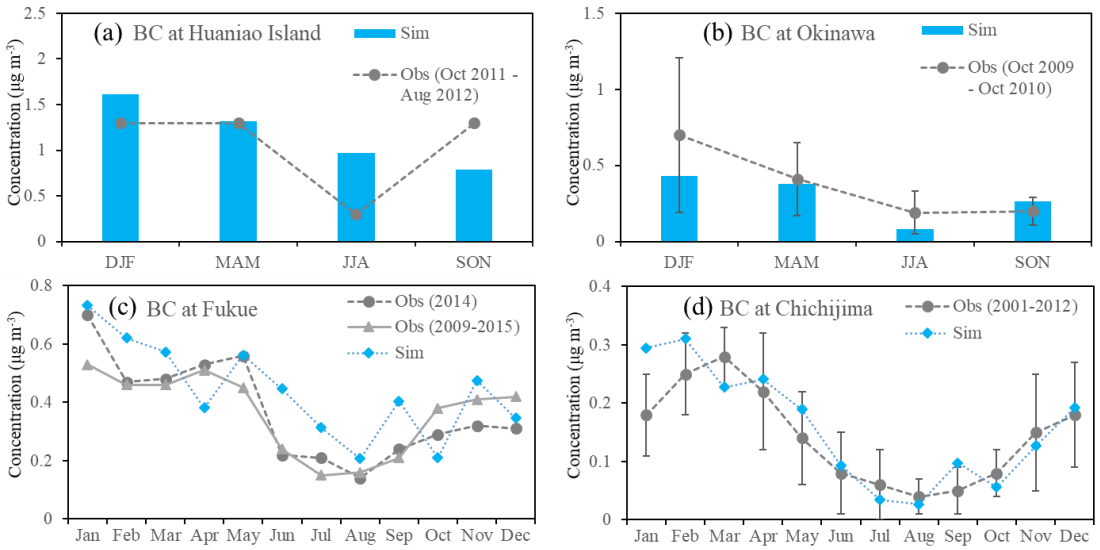


Figure S1. The model simulated (bars) and observed (dotted lines) BC concentrations at different sites. Seasonal mean concentrations were provided at (a) Huaniao Island (Wang et al., 2015) and (b) Okinawa (Kunwar and Kawamura, 2014) while monthly mean concentrations were provided at (c) Fukue (Kanaya et al., 2016) and (d) Chichijima Island (Boreddy et al., 2018). The observed sample standard deviations were available at Okinawa and Chichijima. The simulation is for the year 2014.

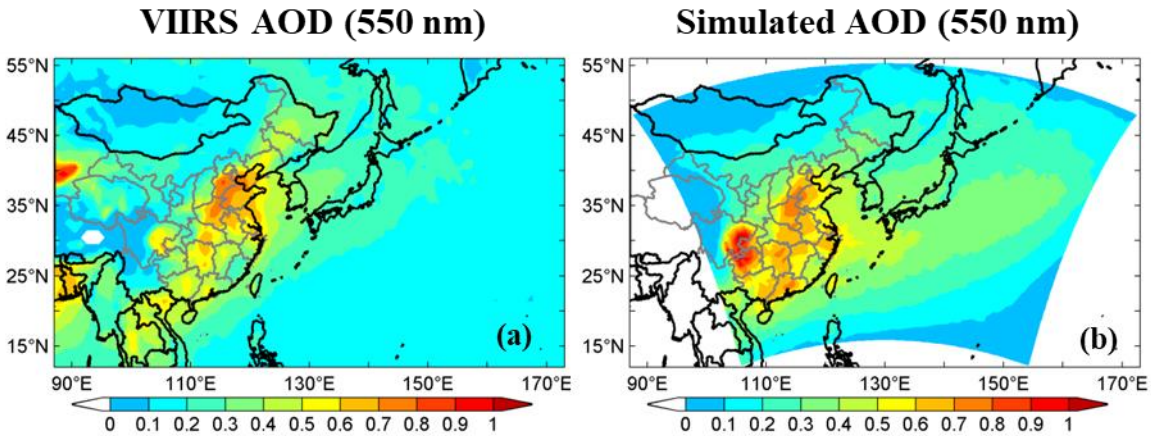


Figure S2. Satellite (VIIRS) retrieved (a) and model simulated (b) annual mean AOD at 550 nm. The model results were sampled according to the satellite retrievals.

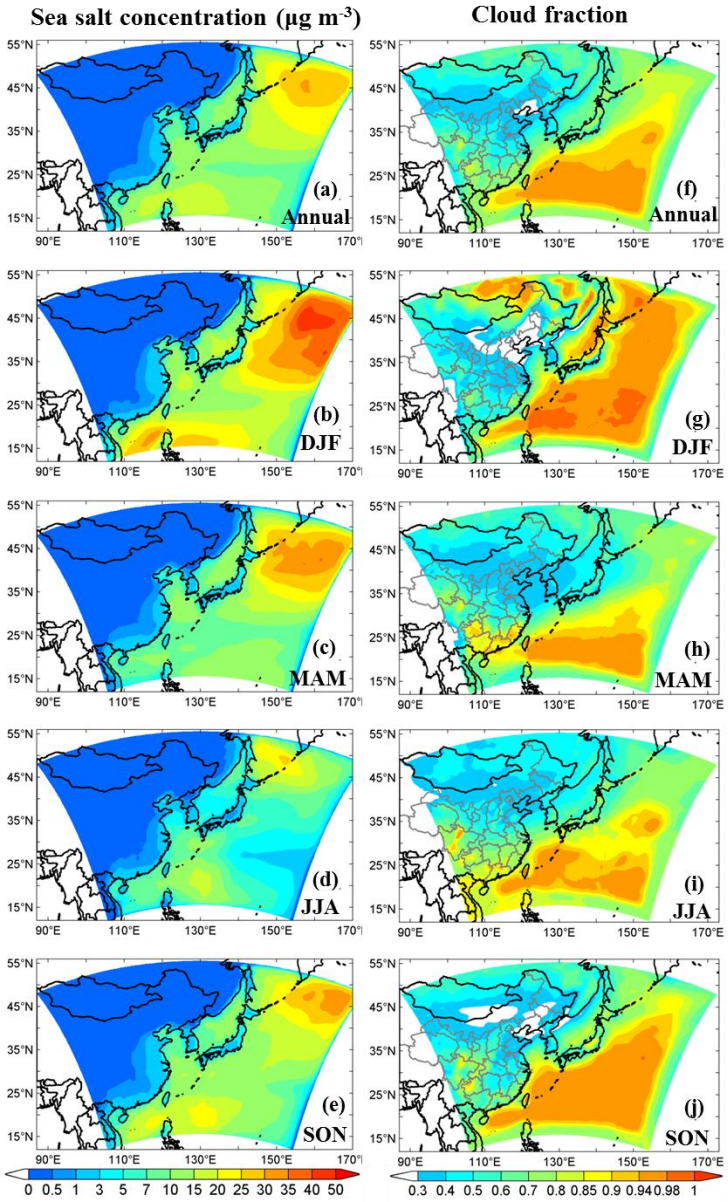


Figure S3. Model simulated annual and seasonal mean near surface sea salt concentrations (unit: μg m^{-3}) (a~e) and cloud fractions (unit: %) (f~j).

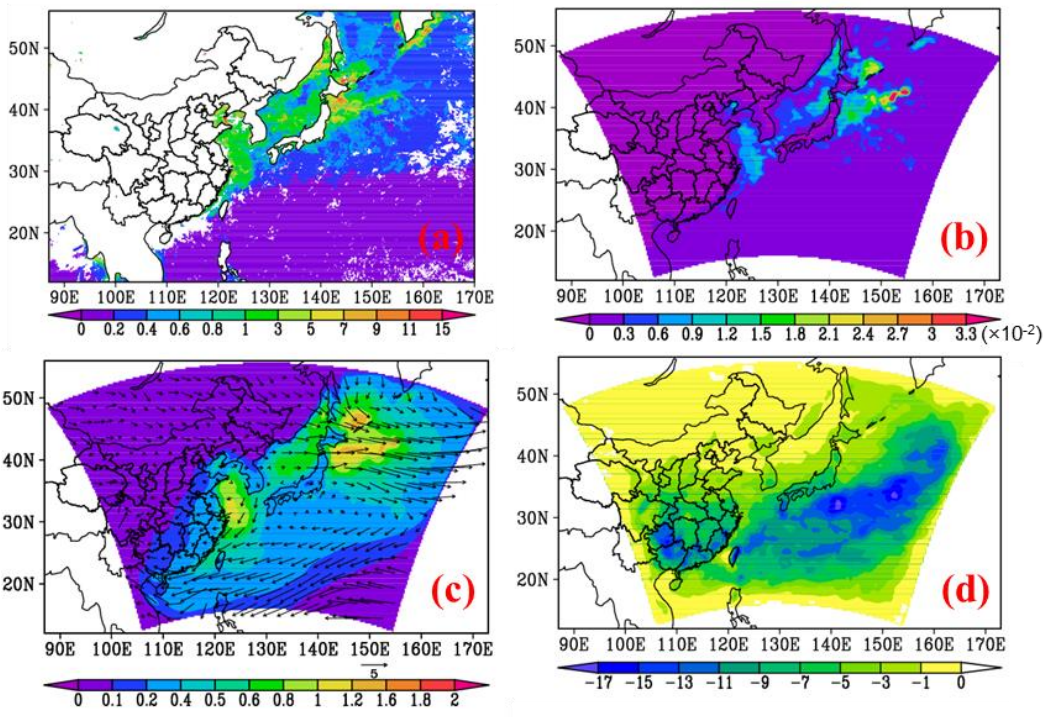


Figure S4. VIIRS retrieved April mean Chl-a concentration (unit: mg m^{-3}) (a), model simulated April mean MPOA emission flux (unit: μg m^{-2} s^{-1}) (b), MOA concentration (unit: μg m^{-3}) overlaid with wind vector (unit: m s^{-1}) (c), and IRE_{MOA} (unit: W m^{-2}) (d).

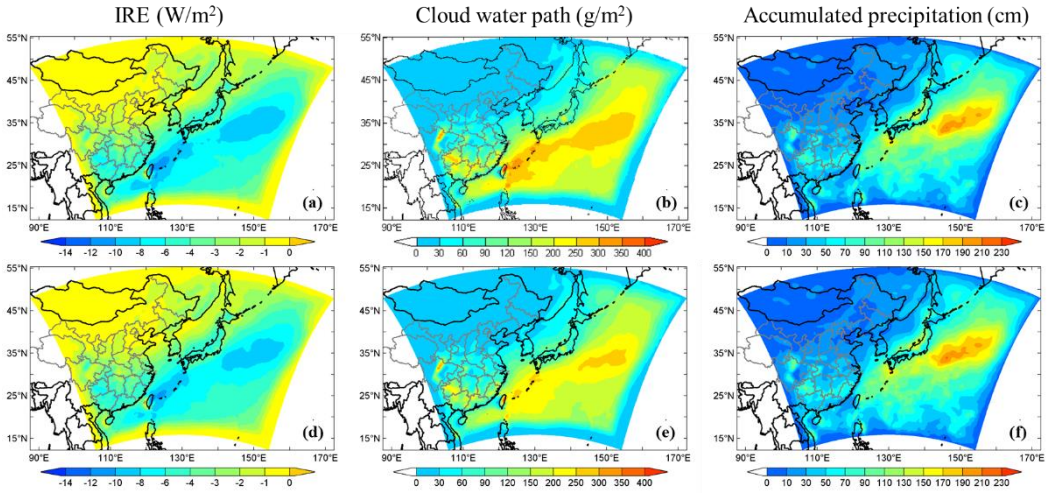


Figure S5. Comparisons between aerosol indirect effect due to MOA from the BASE case (1st + 2nd effects) (a~c) and the sensitivity case with only the 1st indirect effect (d~f). Annual mean IRE (a, d), annual mean integrated cloud water (b, e), and accumulated rain (c, f) are presented.

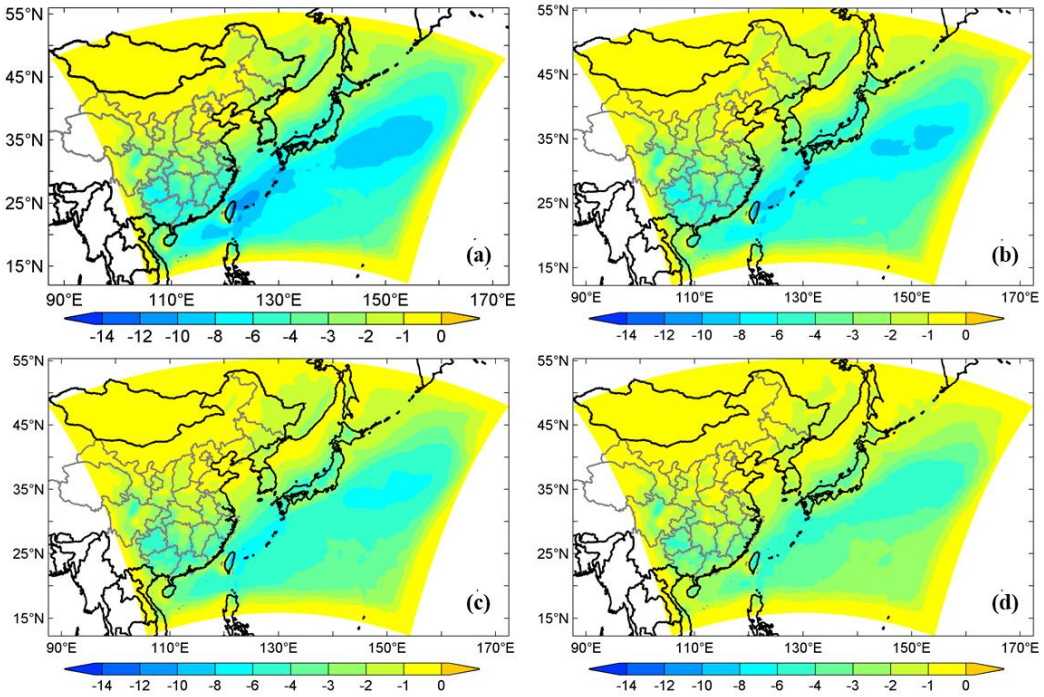


Figure S6. Annual mean IRE due to MOA from the sensitivity simulations. (a) the BASE case, (b) SENS1, (c) SENS2, and (d) SENS3.

Table S1. Observed (Obs) and simulated (Sim) annual mean sodium (Na^+) concentrations at EANET sites (units: μg m^{-3}). Pearson correlation coefficients (R) are presented.

Site	Samples	Obs	Sim	R
Rishiri	13	2.13	3.39	0.75
Tappi	22	2.89	1.84	0.78
Sado-seki	26	2.34	3.77	0.64
Oki	26	3.74	2.31	0.81
Hedo	24	4.79	2.89	0.53
Ogasawara	26	2.54	2.66	0.85
Total	137	3.14	2.78	0.50

Table S2. Performance statistics for hourly gas precursor concentraions (unit: pptv) at EANET sites for the year 2014. Mean observation (Obs), mean simulation (Sim), correlation coefficient (R), and normalized mean bias (NMB in %) are listed.

	SO_2					NO_x/NO_2[a]					O_3				
Sites	Samples	Obs	Sim	R	NMB	Samples	Obs	Sim	R	NMB	Samples	Obs	Sim	R	NMB
Rishiri	8493	0.22	0.21	0.62	-5	7606	0.70	0.96	0.27	38	8583	38.6	45.4	0.57	18
Tappi	8570	0.48	0.40	0.35	-15	8517	1.47	1.95	0.40	33	8569	39.3	48.0	0.52	22
Sado	8233	0.38	0.52	0.41	39	8200	0.75	2.91	0.39	286	8400	46.5	49.6	0.55	7
Oki	7837	0.54	0.45	0.54	-17	7919	1.32	1.66	0.39	26	8507	46.8	50.8	0.52	9
Hedo	7546	0.26	0.26	0.55	-2	7910	0.74	1.10	0.47	48	8260	39.6	41.5	0.84	5
Ogasawara	7635	0.10	0.11	0.09	5	7226	0.37	0.37	0.21	1	8506	33.4	35.8	0.84	7
Jeju	8282	0.54	0.76	0.41	42	8300	3.29	3.53	0.29	7	8419	44.7	52.5	0.56	17
Kanghwa	8517	2.77	4.09	0.25	48	8539	5.90	12.97	0.45	120	8526	50.3	29.4	0.55	-42
Imsil	8337	2.32	1.50	0.16	-35	8200	3.99	1.81	0.18	-55	8385	31.4	39.9	0.54	27
Average	73450	0.86	0.95	0.51	10	72417	2.12	2.87	0.48	36	76155	41.2	43.6	0.54	6

a: NO_x in Japan and NO_2 in Korea.

Table S3. Comparison of observational based estimated marine isoprene emission fluxes over different oceanic areas from previously published studies and model simulations (Units: nmol m^{-2} day^{-1}).

Locations[a]	Simulations from this study		Observations from previous studies		References
	periods	mean (max/min)	periods	mean (max/min)	
Western North Pacific	May 2014	85 (100/40)	18–26 May 2001	140 (300/32)	Matsunaga et al. (2002)
(30.75~35.78°N, 146.42°E)					
Northwest Pacific	Jan 2014	26.2	Jan 2008–2012	21.4	Ooki et al. (2015)
(34~43°N,138~150°E)	May 2014	88.6	May 2008–2012	143.8	
	Aug 2014	63.1	Aug 2008–2012	55.6	
East China Sea	Oct–Nov 2014	48 (120/0)	Oct–Nov 2013	48.34 (169.15/4.19)	Li et al. (2018)
(25.69~30°N, 121~126°E)	May–Jun 2014	35 (180/0)	May–Jun 2014	36.12 (137.75/2.46)	
South Yellow Sea and East China Sea	Jul 2014	130 (240/0)	14 July – 1 Aug 2013	161.5 (537.2/22.17)	Li et al. (2017)
(30~38°N, 121.6~127°E)					
Southern Ocean[b]			Dec 2010–Jan 2011	(313/181)	Kameyama et al. (2014)
Arctic Ocean[b]			Jun-Jul 2010	(148/8.8)	Tran et al. (2013)

a: All the observations were conducted on research cruises and ranges of longitudes and latitudes indicate the coverages of each cruise.

b: Simulation results were restricted within the study domain of western North Pacific.

Table S4. Modeled domain and annual mean IRE due to MOA, cloud water path, and accumulated rain. Model results from the base case (1st + 2nd effects) and the sensitivity case with only the 1st indirect effect are presented.

	Mean[a]	ECS[b]	NWP[c]
	IRE (W/m^2)		
1st+2nd	-4.2	-2.2	-4.1
1st	-4.0	-2.0	-4.0
	Cloud water path (g/m^2)		
1st+2nd	136.7	61.2	142.4
1st	118.5	51.4	124.7
	Accumulated rain (cm)		
1st+2nd	66.2	25.9	94.3
1st	67.1	26.5	94.8

a: Mean over oceanic areas.
b: 27~40°N, 115~123°E.
c: 35~55°N, 140~160°E.

Table S5. Modeled domain and annual/seasonal mean IRE due to MOA from the sensitivity simulations.

	Mean[a]	ECS[b]	NWP[c]
		ANN	
BASE	-4.2	-2.2	-4.1
SENS1	-3.5	-1.6	-3.4
SENS2	-2.8	-1.3	-2.7
SENS3	-2.2	-0.9	-2.1

a: Mean over oceanic areas.
b: 27~40°N, 115~123°E.
c: 35~55°N, 140~160°E.

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