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Comment on acp-2022-394

Anonymous Referee #2

Referee comment on "Contribution of marine biological emissions to gaseous methylamines in the atmosphere: an emission inventory based on satellite data" by Qi Zhang et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-394-RC2, 2022

Overall impression

The study aims to estimate the gas phase concentration of three different amines using WRF-Chem. In addition to previous similar attempts using anthropogenic amine emissions only, the authors apply an online marine biogenic amine emission scheme. This new method seems sound and valid. However, the manuscript lacks of a thorough validation. (Some) Measurement results of different studies are presented in a table but never discussed in context with the simulated concentrations. Furthermore, the conclusions and discussion is not clearly formulated. For example, it is often unclear what type of aggregation / averages are referred to when presenting relative differences. This made it hard to follow the discussion and the results and likely lead to misunderstandings. Finally, potential shortcomings of the current method (e.g. loss to atmospheric aerosols is not considered) is not discussed anywhere in the paper.

Despite the fact that the manuscript could be improved in terms of language and clearer formulations at many points, the manuscript also needs major revision in the interpretation of results, discussion and conclusion. Since measured concentrations were presented, an evaluation section that compares these measurements, in particular the ones of the same year, with the modelled concentrations.

Major comments

l. 138-139: Why is loss to aerosols not considered? Since it was correctly described in the

introduction that amines play a role in aerosol formation, there should be a considerable loss to aerosols. This should be discussed here and in the conclusions.

l. 145-146: It is stated that 2015 was chosen since it had ‚more consecutive days with field observations', however, these field observations were not used in the manuscript for validation purposes. Furthermore, the authors cite e.g. Zheng et al., 2015, in this context, who performed measurements in 2012, so this study probably does not help to explain why 2015 was chosen.

l. 148-149: The mentioned verification discussion is missing in the manuscript.

l. 349-350: Weren't the simulation periods chosen to compare with observations? What were the available observations in the simulated time periods of 2015? Above it was mentioned, that daily measurements are available in certain periods. These periods were simulated. So why don't the authors take the grid cell where the measurement took place and produce a daily mean from the simulation and compare this to the observation?

l. 351-352: Since the range of observed concentrations spans ~an order of magnitude for each of the three investigated amines, it's hard to say that simulation results are 'close'. What you could generally infer is that simulation and available observations agree in terms of order of magnitude. The increase due to MBE does not seem to be very strong seen the numbers in Table 6 to justify the statement 'inclusion of marine biogenic emissions can make up for the shortcomings of the previous models' underestimation of amines'

l. 355: The mentioned verification part is missing. In the text, there are no comparisons

between measured and simulated values and no discussion of the evaluation of the simulated values against the observations shown in Table 6.

l. 364-365: Which region correspond these numbers to? I get different relative changes when using the numbers given in Table 6.

Later I see, that these numbers seem to refer to only regions with an increase shown in Table 7, right? Why do the authors present these numbers and not the domain average in the text (MA in Table 7 if I understood correctly)? In any case, it need to be made much clearer in the whole discussion sections what averages the given numbers in the text refer to. At present it is impossible to follow the discussion of results in a fair amount of time.

Conclusion section: It has to be stated that the drawn conclusions are only valid for the given months in the year 2015. What is with potential limitations since loss to atmospheric aerosol is not considered? This should be discussed. Overall none of the results of the sensitivity simulations in sections 3.3 is summarized in the conclusion. Why not? And a comparison to the available measurements is missing.

l. 484-486: The authors should summarize what they actually did. This numerical modelling to simulate gas phase amine concentration. In addition to anthropogenic emission MBE of amines was treated in an online manner.

Minor comments

Please check the manuscript with a native English speaker if possible. At many instances, some formulations were misleading or seemed at least very unfamiliar.

l. 20: ‚has been insufficiently investigated in the current emission inventory.' What do the author's want to express? An emission inventory cannot investigate.

l. 60-62: Stronger and more easily oxidized than what? Likely ammonia, then please state it like this.

l. 63-65: Is the lower concentration of amines compared to ammonia really a result of faster loss processes only or at least partly due to the lower emission rates?

l. 76-80: It is referred to ‚increased concentrations', an ‚increase by 1-2 orders of magnitude' and a ‚significant increase', but it is not mentioned compared to what concentrations have increased?

l. 89-90: Not sure if China's location alone can be called unique. Densely populated coastal areas can be found in many places in the world. The combination of location and pollution is rather what could be called unique.

l. 95: ‚amines' role in atmospheric chemistry' sounds strange. Better: ‚the role of amines in …'?

l. 101: Is ‚reasonable' the right word? ‚Complete' or ‚detailed' might fit better.

l. 102-103: First, is the emission mass ratio in the Mao et al., 2018, AE emission inventory really ‚arbitrary'? Second, since this scheme is used as it is, and just an online MBE scheme was added, your study does not ‚overcome the arbitrariness' in the AE emission inventory. Please reformulate these sentences in a more clearer way.

l. 104: Multiple -> multiple

l. 121: This is confusing here. Which is the ‚previous method' you are referring to?

l. 127-129: Is an ammonia / amine ratio, e.g. similar to traffic, also applied for ammonia emissions from ships? If not, why not?

l. 150: What do the authors mean with 'representative'? Do these months represent a specific season? If so, is this also the case for these months in the year 2015? As the authors state later, at least SST might be non-typical for parts of the 2015, which indicates it is not representative in general. So, please clarify what is meant with ‚representative' in the context of this paper. I doubt that 10 days of one month can be called ‚representative' for the same month in a climate view or in other years.

l. 154: ‚chemical boundary' -> ‚chemical boundary conditions'

l. 154-155: Did the authors run CAM-CHEM themselves to generate boundary conditions? This is what I understand from reading. However, the webpage given a few sentences later suggest that driving data for boundary conditions can be download. Please clarify.

l. 168: Why the reference to these measurements of concentrations in air and in water by Gibb et al., 1999? How exactly were these used?

l. 188: solvated -> dissolved?

l. 189-190: ‚this study selected the average of the observed values of other sea areas (Table 3)'. I don't understand what the authors want to express here. Which numbers of Table 3 are used in the emission algorithm and how exactly? The description of your method might need more detail or clarification.

l. 198-199: Perhaps related to the previous comment… It would be helpful if the authors can give their calculated average values in Table 3. This helps the reader to much easier follow the method.

l. 208: The unit is probably is 45 g kg-1. The term ‚within' suggest a range but only one number is given.

l. 226-228: Was this done by the authors? If yes, please describe in more detail how the SST data was combined with the other satellite data sets mentioned here. According to the description before, the NESDIS SST data is already on 5 km horizontal resolution.

l. 235: ‚SST presents the seasonality' sounds unfamiliar. Please check for proper English.

l. 257: Obsolete "at the".

l. 275: ion intensity -> ionic strength?

section 2.5.4: Is time-resolved (e.g. hourly) wind speed utilized or monthly mean?

l. 293: Meaning of FNL is missing.

l. 294-295: Well, there is considerable bias for RH. This might not be important for your study, but in any case should not be stated suggestion good agreement. Most importantly is that model and observations of wind speed agree reasonably well.

l. 302: Are the numbers presented in Table 5 for one grid cell or domain mean? Or only over ocean?

l. 303-307: Why not also in table 5 change the signs, which is then consistent with the rest of the figures and the paper?

l. 308, Fig. 3: From table 5 I understand that the ocean is a source for MMA and TMA, but a sink for DMA. Why aren't therefore negative emission fluxes seen for DMA over ocean in Fig. 3?

l. 309, Fig. 4: Is that domain mean?

l. 309: ‚MBE emission' -> emission to much, since the E in MBE already refers to emission.

l. 310, 311: TMA -> ‚MBE flux of TMA' or similar.

l. 309-314: Why no discussion of DMA emission flux, but only for MMA and TMA?

l. 315-335: Just as a suggestion: These paragraphs might be more useful directly after section 2.5, before section 3.1.

l. 315, Fig. 5: Which values were used for the other three input variables, while only one is varied? I understand that the four plots do not show the contribution, but the variation of the emission flux due to the variation of one variable while the others were held constant. It is important for the interpretation and the magnitude of the contribution to provide a clearer description here.

Please also clarify which area or aggregation Fig. 5 refers to. Is that temporal domain mean or ocean mean or one grid cell?

l. 316: ‚Kg, which is directly proportional to Kg' -> Probably the latter Kg is meant to be something else.

l. 325, 338, 344, 346 and later occurrences: overflow -> emission, release or similar.

Check with native speaker.

l. 347: 'affecting the variation of the exchange fluxes' would be more precise in my view.

l. 349, 354: What is the temporal and spatial resolution of the model? This should be mentioned in section 2.3 where the model is described.

l. 358-360: Future tense ('will') does not sound correct.

l. 360-361: No significant change in autumn and winter seems to be the case for MMA and DMA only, according to Table 6.

l. 381 and other occurrences of ‚offshore‘: Shouldn't ‚offshore‘ be on the open ocean, at least farer away from the coast on the ocean? Why is land mass with 500m above sea level considered as ‚offshore’?

l. 393: Formulation sounds incorrect. Perhaps better 'was lowered / decreases by less than 10%'.

l. 396-398: Can that be shown with the model results, e.g. NO, OH concentration maps?

l. 400: What do the authors mean with ‚model mechanism‘?

l. 419-420: ‚they have relatively high fluxes’: What is the relative contribution of residential emissions in the mentioned areas? It seems that this is rather uniform and hence the relative change is more uniform (hence the relative change not as 'obvious' as you say).

l. 420-421: ‚the difference‘ -> If you refer to Fig. S2 and S3, the authors probably mean ‚the relative difference‘.

l. 423: What is high value and low value area?

l. 445-446: Shouldn't the change of pH due to Chla change directly affect the sea air exchange of DMA as well? What do you mean with 'the change of MBE of DMA is not considered'?

l. 447: 'range of DMA' -> 'range of DMA change'

l. 449-450: ‚therefore, DMA reacts with ·OH faster than the other two amines in the upwind direction, and its concentration is less affected.‘ Can the authors please explain in more detail what they mean here?

l. 466-467: ‚going below 50 mm in October’: What region does that refer to? Also in July many places in the domain have precipitation amounts < 50 mm.

l. 475: ‚DMA is only affected by terrestrial AE‘: Didn't it have negative emission fluxes over sea water, i.e. is lost into the ocean surface (Fig. 4)?

l. 476: variation range of its concentration -> variation range of its concentration change

l. 490: It's not the satellite data itself, but its application in an online emission scheme, that might reflect the emission situation.

l. 494-495: Is that 500km away from the coast on land or 500km away over the ocean?

l. 495-496: These are the maximum increases for TMA due to MBE, aren't they? Why don’t authors instead mention the average increases in the text as was done for MMA. I believe that more readers would be interested in averages than in the extremes? Anyhow, it always must be clear what the given numbers are. This is missing throughout the manuscript in many places and makes it rather poor.

l. 499: ‚WS and Chla were found to be the dominant factors affecting MBE fluxes’: This is the case for all amines, right? Then please state it like this.

l. 507: ‚and the ocean also transforms from a source of amines to a sink’: Was that observed in the simulations, since such result was not presented in section 3.3.2? And was it the case for all amines or only some? Since the Chla concentrations, according to Fig. 2, were mostly < 10 mg m-3, 50% increase should only make it a sink for DMA, and very slightly MMA perhaps.

Table 6: Some observation results are presented in µg m-3 and ng m-3. These should be transformed to pptv. Why are the Gao et al. 2022 observations of TMA much larger than for the other studies?

Fig. 2: The numbers in the plots: Black for SST, Chla, SSS and white color for WS would probably give better readability.

Fig. 4: Labels are wrong. In the figure, dashed is MBE and solid lines are AE.

Comments to the author response to the quick initial review comments

In the response, changes in lines 362, 418, 464 are mentioned (with ‚descriptions and data sources'), but I can't find these changes.

The comparison to measurements can of course be done only in a qualitative manner, but it should at least be discussed in the manuscript and not only presented in a table (Table 6).