Interactive comment on “Global ocean dimethyl sulfide climatology estimated from observations and an artificial neural network” by Wei-Lei Wang et al.

Anonymous Referee #2

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The manuscript proposes a new global ocean DMS climatology, or a method to con- struct it, based on an Artificial Neural Network (ANN). This methodology uses a number of variables and their intelligent combinations as predictors of DMS concentration distribution. It is meant to overcome the limitations of objective analysis based on inter- and extrapolations as well as the limitations of simple linear or logarithmic regressions with few predictors, and to provide better fits of predictions to observations. While developing their ANN application and to claim its better performance, the authors conduct parallel applications of previously published models. Eventually, they indeed obtain a better fit, but very similar seasonal and geographic distributions. The global annual emission to the atmosphere is revised towards the lower end of the hitherto most accepted estimate.

The topic is timely since, after years of having DMS been dismissed for its role in new particle formation, recent studies are recognizing it again as a central agent in ocean- atmosphere-climate interactions. Atmospheric chemistry and climate models require updated climatologies of DMS emissions.

Thank you for your positive comments.

The text is generally well written and the display items are clear and informative, with one exception (see particulars below).

That said, the manuscript reads as though it was written 10 years ago. Even though the ANN methodology is probably state-of-the-art (I am not an expert and can hardly assess every technical aspect), the interpretation arguments are outdated, ignoring many of the discoveries in the last decade. This adds to some bad referencing. But most importantly, when the authors intend to make relevant comparisons with previous similar efforts, they miss the point of the studies they are comparing to, or use them in the wrong way. Finally, besides presenting their new method, they fail to discuss what is new in their findings, they just repeat what is already well known and with much poorer arguments, rather than stressing what is unveiled and why. I will develop these and other concerns hereafter, as they come up in the order of the manuscript.

L28-30: “The weak relationship may be caused by the so-called “summer DMS para- dox”, which describes a phenomenon where a maximum DMS concentration is commonly detected in low latitude waters when phytoplankton biomass is low (Toole and Siegel, 2004; Vallina et al., 2008).” This is not the summer DMS paradox (a term, by the way, suggested by Simo & Pedros-Alio Nature 1999), which states that the annual maximum of surface DMS commonly occurs in summer, even at the mid and subtropical latitudes where chlorophyll-a (chl-a) is at its annual minimum.

Thank you for your correction.

We have rephrased the statement and added corresponding reference as follows,

“The weak relationship may be caused by the so-called “summer DMS paradox”, which describes a phenomenon that annual maximum of surface DMS concentration is commonly

detected in summer when Chl *a* is at its annual minimum in mid and subtropical low latitude waters (Simó and Pedrós-Alió, 1999)."

L34-35: "Simó and Dachs (2002) achieved a strong relationship between heavily binned and averaged DMS data and mixed layer depth (MLD)." This is not true. Simo & Dachs (2002) correlated DMS to the MLD and to chl-a/MLD, depending on a chl-a/MLD threshold.

We have corrected the corresponding statement as follows:

"Simó and Dachs (2002) achieved a strong linear relationship between heavily binned/averaged DMS and mixed layer depth (MLD) when Chl-a/MLD ≥ 0.02, and a logarithmic relationship between DMS and Chl-a/MLD when Chl-a/MLD < 0.02."

L53-54: "Many provinces lacked adequate data to create a reliable climatology (Fig. A1). In those situations, temporal interpolations were used to fill the blanks, and to create a first-guess map." This was done where monthly data gaps existed to complete the seasonality. Where data were lacking to even outline a seasonality, this was taken from a neighboring province and adjusted to the existing data.

Thank you for pointing this out. We have adjusted the description as follows,

"Many provinces lacked adequate data to create a reliable climatology (Fig. A1). In those situations, they first generated an annual cycle with monthly means for each province. Temporal interpolations were used to fill the monthly gaps if there were enough data to create a robust annual mean. Otherwise, interpolation from neighboring provinces was used to fill the remaining gaps."

L61: "Since DMS is produced by marine that algae. . ." This is totally outdated. There are tens of papers showing that this is an oversimplification. DMSP is mainly produced by marine algae, and it is transformed into DMS by marine algae, bacteria and with involvement of zooplankton.

We have rephrased the statement as follows,

"The precursor of DMS, DMSP, is mainly produced by marine algae (e.g. Kiene et al., 2000; Curson et al., 2011), and a small fraction of DMSP is transformed to DMS by marine algae and/or bacteria lyases (Simó, 2001; Stefels et al., 2007; Curson et al., 2011; Moran et al., 2012), and mostly as a result of food web interactions (Kiene et al., 2000; Simó, 2001)."

L93-94: "We do not log-transform SST to avoid losing data with temperature below (equal to) zero." You may have other reasons to not log transform SST, but not this one. A common practice to log transform SST if desired is to convert it to K (Kelvin) first.

We have re-run the model using log-transformed K.

If I understand it correctly, you use chl-a data where available, otherwise you take it from SeaWiFS. What efforts have you done to reconcile in situ with satellite chl- a? It is well known

that algorithms for satellite estimates of chl-a are developed and calibrated against HPLC chl-a, and there is an important shift between this and Turner fluorometric chl-a. Therefore, putting together in situ (Turner, perhaps HPLC too?) and satellite chl-a data will mess up your statistics.

Thank you for pointing this out. In the revised MS, we used only satellite Chl-*a*, and added more discussion as follows (l.84 – 1.92),

"SeaWiFS Chl-*a* data (Level 3-binned, spatial resolution of 9.2 km) from December 1997 to March 2010 were matched to DMS data according to coordinates and sampling date. We compared PMEL in situ Chl *a* to SeaWiFS Chl *a*, which are well correlated on logarithmic scale (R^2 = 0.64) with a slope of 0.67 and an intercept of -0.06, $[\log(Chl_{SeaWiFS}) = 0.67\log(Chl_{in-situ}) - 0.01]$, which means that on logarithmic scale SeaWiFS Chl-*a* concentrations are on average ~30% lower than those of in situ Chl-*a* concentrations. This is possibly because SeaWiFS Chl *a* is calibrated based on HPLC determined Chl *a* (Morel et al., 2007), which on average is ~40% lower than that determined using Fluorometric method (Sathyendranath et al., 2009). Unfortunately, there is no flag in the database showing how Chl *a* was determined. For consistency, we use only Chl-*a* data retrieved from SeaWiFS in the following multilinear and network models."

Calculation of air-sea fluxes: I agree that Nightingale 2000 is quite a standard. But, why not using a more updated linear relationship of Kw to u10? Marandino proposed one with one of the coauthors. Also, you use monthly means of wind speed. Since you are using a nonlinear dependence of Kw on the u10, how do you deal with the fact that a mean u10 will not give the same result as a mean Kw?

In the paper we used two flux parameterizations, GM12 according to Goddijn-Murphy et al .2012 and N00 according to Nightingale et al 2000. GM12 DOES describe a linear relationship between Kw and u10, and it is more updated that Marandino et al 2008 (if this is the reference that you referred to).

We also used N00, because we can compare to previous results that used the same parameterization.

We did a correction on mean u10 as described below (l.209 – l.214),

"Because the N00 parameterization was calibrated using in situ wind speeds and has a nonlinear quadratic dependence on wind speed, the use of monthly mean wind speeds will introduce errors. To reconcile differences between in situ wind speed and monthly mean wind speed, a correction is applied according to Simó and Dachs (2002) by assuming that instantaneous wind speeds follow a Rayleigh distribution. Eq. 8 thus becomes $k_{w,660}$ $=[0.222\eta^2\Gamma(1+2/\xi)+0.333\eta\Gamma(s)](ScDMS/600)^{-0.5}$, where $\eta^2 = 4U_{10}^2/\pi$; $s=(1+1/\xi)$, and $\xi = 2$ for Rayleigh distribution (Livingstone and Imboden, 1993)."

L170-176: It reads as though you did not know of the existence of Gali et al. BGS 2016.

We have added the following discussion (l.226 – l.229),

"McParland and Levine (2019) developed a mechanistic model that related intracellular DMSP concentration to environmental stress, and coupled the model with MIT ecosystem model (DARWIN) to estimate global ocean DMSP distribution. Galí et al. (2015) first applied a remote sensing algorithm to obtain a DMSP climatology, from which they predict DMS climatology through an empirical relationship with PAR (Galí et al., 2018)."

L182: "On the other hand, negative correlations between DMS and Chl a have also been detected in coastal waters of the Mediterranean and in the Sargasso Sea (Toole and Siegel, 2004)." Toole & Siegel did not do anything with Med Sea data. The original data from the Sargasso Sea were from Dacey et al DSR 1996, and data from the coastal Med Sea were reported by Vila-Costa et al. LO 2008.

Thank you for pointing this out.
We realized that it is that Dacey et al. (1998) reported the original Sargasso Sea data, and Toole and Siegel analyzed the correlation between DMS and Chl a.
We have added the original reference and changed the wording.

"On the other hand, there are numerous studies that failed to correlate DMS and Chl a (e.g. Dacey et al., 1998; Kettle et al., 1999; Toole and Siegel, 2004)."

L185-190: This is a very poor interpretation of the DMS vs MLD coupling, and a misuse of the original relationship suggested by Simo & Dachs GBC 2002. As a matter of fact, you cite Simo & Pedros-Alio GBC 1999 because they brought it up for the first time, but the occurrence of a negative relationship between DMS and MLD over large regions of the global ocean was reported by Simo & Dachs. However, the relationship was logarithmic, DMS = a*Ln(MLD) + b, and there are reasons for this to occur, related to exposure to solar radiation. Trying to correlate DMS directly to MLD (or in a log-log manner) is not expected to provide good prediction.

Thank you for pointing this out. We have added more discussion as follows,

"It is proposed that stratified (high light) environment nourishes strong DMSP producers, or phytoplankton cellular DMSP quota increases in such an environment. High conversion rate from DMSP to DMS in stratified waters is another reason for high DMS concentrations when MLD is shallow. Meanwhile, the biological DMS consumption rate decreases in oligotrophic oceans (Galí and Simó, 2015). Dilution model, which describes a phenomenon that when the mixed layer deepens water with no or little DMS is entrained into the surface waters and dilutes surface DMS concentrations, was also proposed to explain the anti-correlation between DMS concentration and MLD (Aranami and Tsunogai, 2004). However, recent studies show that DMS loss rate via vertical mixing is orders of magnitude lower than production/consumption rates (e.g. Galí et al., 2013c; Royer et al., 2016)."

L189-199: There are a number of papers that should be cited here – besides Toole et al. and Sunda et al, several papers by Marti Gali deal exactly with the effects of solar radiation, and particularly UV, on enhancing DMS production and concentration.

"Climatological PAR is the second strongest predictor (R2 = 0.12, n = 54,683) of raw DMS data with a positive correlation. (. . .) Strong correlation between monthly binned and averaged solar radiation dose (SRD) and DMS concentration has been reported (R2 = 0.94) at the Blanes Bay Microbial Observatory located in the coast of northwest Mediterranean (Vallina and Simó, 2007)." Again, you compare your statistics with that of a previous study, but applying a different calculation. According to the methods description, you used monthly PAR, i.e., monthly surface irradiance. Vallina & Simo 2007, conversely, computed what they called the solar radiation dose, which is the daily averaged solar radiation integral in the mixed layer. This is very different from surface irradiance, because it takes into account the mixed layer depth (and a median light attenuation coefficient). Later on, in L200-211, you infer that, contrasting to Vallina & Simo, you did not get a good correlation to light, and attribute it to the number of original data and to data binning. But you did not use the same light metrics as the other authors, and ignored the arguments given by V&S to use the SRD instead of the surface irradiance, and ignoring the Gali & Simo GBC 2015 meta-analysis too.

Good points.
We have rebuilt the SRD vs DMS model, recomputed the results, and revised our discussion as follows,

"Vallina and Simó (2007) reported an R^2 of 0.95 (n=14) between DMS concentration and SRD. We applied the same linear regressions on both raw data and monthly 1°× 1° data, and found no significant correlations between DMS and SRD that is calculated according to Vallina and Simó (2007) using the following equation,

$$SRD = SI \cdot \frac{1}{Kd490 \cdot MLD}(1 - \exp(-Kd490 \cdot MLD))$$

where SI is shortwave irradiance (W m^{-2}), which is converted from PAR according to Galí and Simó (2015)."
L201: "Simó and Dachs (2002) obtained a high R2 value between DMS concentration and the ratio of Chl a and MLD (Chl/MLD)." This is not true. As already mentioned above, the Simo & Dachs (2002) model correlated DMS to the MLD (logarithmic) and to chl-a/MLD (linear), depending on a chl-a/MLD threshold.
Yes, we used exactly the same model, and have made it clear in the text as follows (l.319 – l.329),

"Simó and Dachs (2002) obtained high R^2 values between DMS concentration and the ratio of Chl a to MLD (Chl/MLD) when Chl/MLD is greater than or equal to 0.02, and between DMS concentration and ln(MLD) when Chl/MLD is less than 0.02. We tried exactly the same model on raw PMEL data with in situ Chl-a measurements and climatological MLD, and found that both correlations between DMS and Chl/MLD (n = 4,921, R^2 =~ 0 .1) and between DMS and ln(MLD) (n = 5,978, R^2 =~ 0) are statistically insignificant. To reduce interannual variability, we binned in situ Chl a and DMS into monthly 1° × 1° grid, and retested the above model. We found that the correlations are still statistically insignificant. (R^2=~ 0)"
."

All in all, if you are to compare your statistics to those of S&D 2002 and V&S 2007, everything here has to be recomputed and rewritten.

The results were recomputed and the text was rewritten. Please see our responses to your above comments.

The arguments against binning the data are poor. It is true that binning reduces the variance, but you are using monthly climatologies (heavily averaged and also binned) to relate raw DMS data to potential predictors. Also, binning must be used if you want to avoid giving too much predictive weight to the regions thoroughly sampled over the undersampled. This is becoming more important as we are bringing in underway data at unprecedented spatial resolution, like the NAAMES data incorporated here.

Since this issue was brought up by all three reviewers, we have added adequate discussion/arguments to this point as shown in the following text.

The PMEL database expanded dramatically. Now there are a total of 86,785 valid DMS measurements (concentration greater than 0.1 nM and less than 100 nM according to your instructions), that is 71% larger than the number of data we initially used (51,161). For the expanded data set, ~93% of DMS are accompanied with in-situ SST measurements, ~81% are accompanied with in-situ salinity measurements. More importantly, each data point has their unique location and sampling time signatures. As shown in the following figure, sampling time (date) and location information is a strong DMS predictor, which together can decrease DMS root mean square error to 0.64 (on natural logarithm scale). Adding other climatological predictors can further improve the model performance.

The NAAMES dataset has 6,786 valid data points, which are ~7% of the total data points (93,571 = 86785+6786). All data are accompanied with in-situ Chl a, SST, and SAL measurements. For parameters without in-situ measurements, high resolution data are used to match DMS measurements, 0.0417° ×0.0417° for PAR, 0.5° ×0.5° for MLD, and 1° ×1° for NO3, which ensures most of DMS have a set of unique predictors. As shown in Table 1, merging NAAMES data with PMEL data does not significantly change the statistic.

Moreover, binning the data will reduce data variance, which has been demonstrated by Derevianko et al. (2009). The objective of this study is to train an ANN with as many data as possible, so that the model is generalized. It not only can apply to coarse resolution predictor fields, but also can apply to very fine resolution field, for example, we have applied the network to fine resolution NAAMES fields for comparison with in-situ DMS measurements (Bell et al., in prep.).

Lastly, binning data will also result in loss of information. A great amount of information is associated with sampling time and date as shown in the following figure (Fig. 2a in MS). By binning the data into monthly 1° × 1° grid, the valid DMS data points will decrease significantly from 82,996 to 9,018; sampling date feature (365) will be average to 12 months, and coordination combinations will be averaged from 87,332×87,332 to 180°×360°, which

represents great information reduction. For ANN models, less data points usually lead to overfitting Fig. 2b.

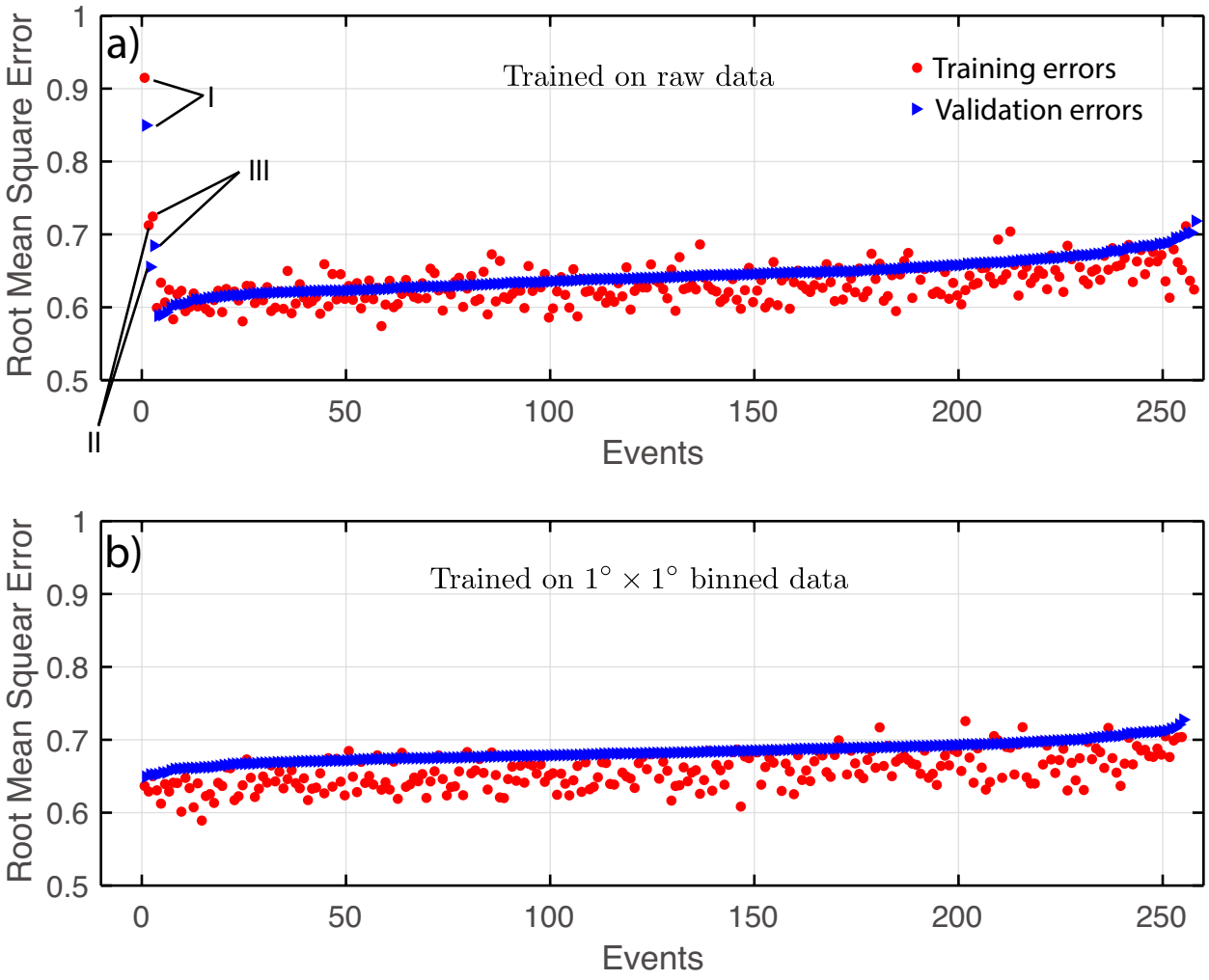

Fig. 2 Parameter sensitivity tests on raw and binned data. (a) Root mean square error on logarithmic scale for the model trained using raw data; (b) Root mean square error on logarithmic scale for the model trained using binned data. The time and location parameters are tested separately without combining with environmental parameters as shown in the upper panel, (I) with only location parameters; (II) with location and day of year parameters; and (III) with location, day of year, and time of day parameters. The model with three location parameters (I) has a root mean square error on natural logarithmic scale of ~0.83, which decreases to ~0.65 by adding sampling day of year parameters (II), however, increases to ~0.67 by adding sampling time parameters (III). We, therefore, do not include sampling time parameters in the following tests. We tested every possible combination of the eight parameters (PAR, MLD, SST, SAL, Chl a, DIP, DIN, and SiO), which in total are 255 tests.

L231-233: “The summertime high DMS concentration at high latitudes is consistent with the hypothesis that phytoplankton use DMSP as a cryoprotectant (Karsten et al., 1992). It is found that the same phytoplankton (Antarctic macroalga) contains higher DMSP concentration in the polar regions than in the temperate regions (Karsten et al., 1990).” Poor again, if not wrong. See recent papers on DMS in polar regions (e.g. Webb et al Sci Rep 2018, Gali et al. PNAS 2019). And macroalgae are not phytoplankton.
Good points. We have updated the reference, and added more discussion as follows,

Good point. We have updated the reference, and added more discussion as follows (l.371 – l.376),

“The summertime high DMS concentration at high latitudes is believed to be linked to the release of ice algae that are prolific DMSP producers (Stefels et al., 2012; Webb et al., 2019). As an important cryoprotectant and osmolyte, DMSP helps ice algae to cope with the low temperature and high salinity conditions (Thomas and Dieckmann, 2002). High DMS concentrations at high latitudes have also been observed to accompany blooms of coccolithophores and Phaeocystis, which are strong DMSP producers (Neukermans et al., 2018; Wang et al., 2015). The shoaling of mixed layer depth in summer provides favorable conditions, i.e. stable and warm, with adequate irradiation for coccolithophores and Phaeocystis growth (Galí et al., 2019)”

Subsequent discussion: The seasonality and geographic distribution of DMS have been profusely (and much better) discussed by Lana et al. GBC 2011 and others, including regional studies. You should rather focus on new features unveiled with respect to others, particularly Lana 2011.

In the revised MS, we have shortened the spatial distribution discussion, and added more discussion on the comparison with previous results.

L305-306: “By contrast, objective interpolation methods are spatial/temporal averages of sparse data with no underlying basis in environmental variability.“ Again, this is not totally true. In Lana et al. 2011, to create a first guess field, biogeographic provinces were used, which is an informed approach to extrapolation. These provinces are defined from environmental descriptors. And a distance weighted interpolation from original data was used for interpolation.

We agree that the provinces are defined from environmental descriptors, however, we also noticed that the provinces are static with no seasonality. In the revised version, we weakened our expression as follows,

“By contrast, objective interpolation methods are spatial/temporal averages of sparse data with weak underlying basis in environmental variability.”

Figure 2: An annual average is not very informative. I would even argue it is misleading in the case of highly seasonal variables like DMS, because summer maxs and winter mins cancel out

each other. I would recommend splitting the map into two or four seasons to show hemispherical patterns.

Good point. We split the global map into Southern and Northern hemispheres, and plotted seasonal cycles for each hemisphere (Fig. 3). Also, we made zonally mean average for each season (Fig. 4).

Figure 4: Some differences are outstanding but you do not discuss them. For instance, Lana 2011 captures the September max of DMS concentration in the subarctic NE Pacific, because it is well covered with data. Conversely, your ANN does not capture it. This warrants some discussion, as it will reveal some of the caveats of the ANN approach.

This is an interesting point.

Based on Fig. A1, the observational data do not show high DMS hot spots/or good coverage in September in the subarctic NE Pacific, instead, there are some high DMS measurements in August in this region. Both Lana et al, 2011 and our ANN model capture the high DMS concentrations in August in this region.

We believe that in September the high DMS concentrations in this region are interpolated from August. On the other hand, our ANN model predicts moderate DMS concentration at this region in September.

In any event, we agree that more discussion is needed here. The following discussion was added to address the methodology difference between objective interpolation method and ANN method (l.353 – l.364).

"L11 stands out in the S. hemisphere monthly mean plot (Fig. 3b), with the highest mean concentrations in January and December, when DMS concentrations are ~2 times higher than other model predictions. Galí et al. (2018) identified five short-comings associated with the direct interpolation method employed by Lana et al. (2011). All shortcomings concern the nature of in situ DMS data, including right-skewed distribution, lack of spatial and temporal coverage, lack of duplicate measure- ments, and sampling bias towards DMS-productive conditions. Because of the sparsity and skewed distribution, the interpo- lation/extrapolation method broadcasts small scale features to large scales (Tesdal et al., 2016). This is especially true for the month of January and December when the elevated L11 monthly means were mainly driven by a small amount of extremely high DMS measurements (>40 nM) near the Antarctic continent. On the other hand, empirical models including the ANN model used in this study rely on environmental parameter climatologies to obtain the DMS climatology. Extreme conditions are smoothed out in climatological data, e.g. in the DMS database the maximum in situ Chl-a concentration is > 800 mg/m3, whereas it is ~50 mg/m3 in the SeaWiFS climatology. When climatological data are used to generate DMS distribution, a smaller variance than in situ data is expected."

In summary, I think that the ANN is an interesting approach that will help improve DMS (and other) climatologies, especially where data are lacking, as it will do better than inter- and extrapolations. However, the present manuscript does not go much beyond the application of the ANN; when it intends to do so, too often it uses the wrong arguments and is not fair with previous studies. It fails to mobilize what we have learned about DMS in the last one or two decades.

In the revised MS, we have added more tests and more discussion accordingly. More specifically, we designed experiments that help to open the network “black box”, and based on the results, we thoroughly discussed the parameters that exert an impact of the prediction ability of the model. The text is from line 253-line 303.