Response to Anonymous Referee #1

Comment

This paper presents a global synthesis of Holocene dust records in various archives and provides an updated dataset including the dust mass accumulation rate (DMAR) and particle-size distribution (PSD). The compiled DMAR record was also compared with the modeled variability of global DMAR record. This topic is quite ambitious since numerous dust records were available with great uncertainties. Nonetheless, this paper tried to select those high-to-medium quality data from land, ocean and ice based on several criteria and provided a global compilation of the Holocene DMAR records. Thus, I think that this work is suitable for publication in "Climate of the Past".

Response

Thank you for your valuable comments on our manuscript.

Comment

However, based on the relatively concise content of the main text (e.g., section 4), It remains difficulty to reach some consensus regarding the Holocene dust cycles in different archives/regions, for example, the differences of the DMAR and PSD between Mid-Holocene (6ka) and late Holocene (PI), and between Mid-Holocene and LGM for reference. Given the great authorship, the readers might expect to see a comprehensive review on the dust records on several representative archives (e.g., Antarctica ice cores, Tropical Atlantic marine sediments, and Chinese loess deposits).

Response

We agree that placing in context the dust records with the climate conditions of the different regions, also by comparing to other paleoclimate proxies, is indeed a necessary step in understanding the dust records. Note however that while our work provides the tools for relating DMARs and climate, such a detailed and conclusive analysis for each of the regions is beyond the scopes of our paper, also given different possible interpretations of the multiple proxies presented in the existing literature. The same holds for the interpretation of dust size distributions. We see this work more oriented at providing a methodology and highlighting a few clear and central issues related to the dust records, which was already an ambitious undertaking as the reviewers commented. Nonetheless we welcome your suggestion and we will recommend that future work by ourselves and others should devote proper attention to this aspect for specific regions. The discussion in dust size distributions has been expanded – see comment below.

Comment

Unfortunately, the summary is relatively simple, for example only two paragraphs mentioned about Chinese loess and many references with good OSL dates for the Holocene soil were not fully cited (e.g., Holocene work with OSL dates by Lai and Wintle (2006), Lu et al (2007), Sun et al (2012), Kang et al (2013)).

Response

We appreciate your suggestions for additional OSL dates that were not mentioned in our manuscript. We incorporated the two records among those that fit our selection criteria as described in the manuscript: Weinan and Jingyuan. All the figures and model-based reconstructions were updated based on the expanded dataset.

Comment

Besides, this paper tried to consider the dust PSD, but I cannot find any curves to show either the distribution pattern of dust particles or the time series of dust PSD variations.

Response

Excellent point. We expanded the discussion on particle size distributions, by adding the new Section 4.13 and Figure 18, as well as some final consideration in the conclusions. Please refer to the revised manuscript.

Comment

From Fig.6 to Fig.15, I wonder whether it's possible to add the age controls and errors of these MAR estimations. **Response**

We agree this is a good idea: in fact this is done in the "descriptive sheets" of the Supplement for each record. We avoid adding this piece of information in the aforementioned figures for readability, but we refer to the supplement in the figure caption so that readers are reminded to use that resource: "Please refer to the descriptive sheets in the Supplement for a graphical display of the uncertainties for each record"

Comment

Based on these MAR curves, it's hard to judge the relationship between DMAR and climate changes. It might be perfect to incorporate the typical climate records from different region/archives in these curves, for example, d18O of ice core, benthic d18O of marine records, and magnetic susceptibility of loess deposits.

Response

As we explained in the answer above, this is beyond the scope of this paper. We strongly agree that future work should address those issues for specific regions. We added this consideration to our conclusions: "The work presented in this paper provides the tools for relating DMARs and climate; future work will need to place in context the dust records with the climate conditions of the different regions, also by comparing to other paleoclimate proxies."

Comment

Finally, Fig.17 displays the dust deposition flux variability from the CESM during different time slices, however, the corresponding external forcing and internal boundary conditions in these time slices were not clearly expressed in the text.

Response

The description of the modeling setup is described in Section 3.5 of the Methods. As we explain there, the climate conditions are those of the Mid-Holocene (6 ka BP) and pre-industrial (1850 AD) climates, according to the CMIP5/PMIP3 prescriptions in terms of greenhouse gases concentrations, orbital forcing, and surface conditions, i.e. vegetation cover, ice sheets, sea level (the latter two not relevant for these time slices). The initial conditions were taken from CMIP5/PMIP3 simulations with the CESM. The prognostic dust cycle was scaled according to the methodology outlined there, i.e. based on the observational data.

Response to Anonymous Referee #2

Comment

This paper aims to provide a global synthesis of Holocene dust mass accumulation rates (DMAR) and particle size distributions (PSD) from various archives. Additionally the compiled data is compared to a global model of the dust cycle using the Community Earth System Model (CESM).

S. Albani et al. present a transparent process which they use to select DMAR time series from various archives ranging from marine over lacustrine and terrestial sediments to ice cores. Considering the great uncertainties associated with the individual archives and methods and the differences between the archives this compilation is an ambitious undertaking. It demands a careful review of each type of archive and each individual record which is the major part of the presented work.

The manuscript fits well into the focus of "Climate of the Past" and is in general suitable for publication. However some minor clarifications and changes are needed before final publication.

Response

Dear Referee #2,

Thank you for your detailed comments and suggestions, which we incorporated in our revised version of the manuscript. See our point-to-point response to your comments below.

Comment: Difference to DIRTMAP

The presented study is similar to the DIRTMAP effort initiated by Kohfeld and Harrison (2001). S. Albani et al. provide some comparison of their work to DIRTMAP, however, it would be very helpful for the reader if the differences where stated more clearly within the introduction.

Response

We added a paragraph in the introduction to make explicit from the beginning of our paper the main innovations of our work compared to DIRTMAP: "Inspired by DIRTMAP, our new compilation considers DMARs as the key variable for a coherent study of paleodust archives. The elements of innovation that we introduce here (size distributions, temporal resolution, and attribution of confidence level) however constitute a leap forward into a new generation dust database."

Comment: Terminology

The terminology used in this study to describe the different parts that make up an eolian dust archive are very much specific to sediment archives and is not applicable to ice cores even though the concepts might be the same. It would be desirable to use archive agnostic terms (e.g. Matrix or Archive Accumulation Rate) such that it is immediately clear that the concepts are valid for all the records presented here. However, I am not sure about the exact terms to use. In any case, some additional sentences are needed within the introduction to clarify the transfer of the different terms to non sediment archives.

Response

In our manuscript we used the term "sediment" and derivatives in a loose sense that also encompasses ice, in addition to other sediments *sensu stricto*. We make this explicit in the text in order to avoid confusions, as suggested, at the beginning of Section 2: "Throughout the paper we use the term "sediment" in a broad sense that encompasses ice as well as other sediments in a strict sense."

Comment: PSDs – Section 3.4

S. Albani et al. put great stress on the importance of the particulate size distributions for the usability of the archives. They use a simple rebinning approach to facilitate the intercomparison of the size distributions within in the database. The approach is well described in this section of the manuscript, however it remains unclear whether number or volume distributions where used and are published in the database. Both of these informations should be added.

Additionally one very important information gets lost through the rebinning: the upper and lower limit of the dataset. Due to analytical constraints only part of the complete size distribution is usually observed. The resulting truncation of small and large particles can have a big influence on the total DMAR depending on the truncation

limits. In the compiled database, bins below and above the limit are given as containing zero (counts/volume), when they should be reported as missing values. There is a huge difference between not observing anything and observing nothing. For that reason, the detection limits should be stated alongside the size distributions. S. Albani et al. also do not clearly state how they deal with the truncation in the derivation of the total DMAR and

S. Albani et al. also do not clearly state how they deal with the truncation in the derivation of the total DMAR and how this translates into the size bins used for the model exercise.

Response

Excellent points. We clarify that we always refer to volume size distributions: "We always refer to volume size distributions, both in the main text and the Supplement."

We welcome the comment on clearly addressing the size range of the measuring device. We updated the tables of database by replacing zeros with missing values as appropriate.

The truncation of the size distributions to 10 μ m diameter to match the model's size range is based on the new binning scheme. Similarly the comparison of data versus model size distributions that we introduce in the revised version of the manuscript (Fig. 18), is also based on clustering the binned size distributions in order to match the model bins. We clarify this aspect in in the new Section 4.13 and related Figure 18, as well as in Section 3.5: "...limited to the model's size range i.e. <10 μ m: we considered only the relevant fine fraction from the new binning."

Comment: Fine dust fraction – Table 1

For some of the archives used by the authors no information about the particle sizes are available. For these, S. Albani et al. provide the fine fraction (<10 μ m) seemingly arbitrary in Table 1 of the manuscript. It remains entirely unclear whether these fractions were given by the original authors of the studies associated with the records or if they where set by the authors of this study.

Given that these estimates of the fine fraction are an important part of the comparison between model and data the source and/or approach used should be stated very clearly in the text.

Response

We report a clarified explanation in the caption of Table 1. We added: "Reference to the original studies is provided in the second column from the right. The rightmost column reports the details of how the percentage of $DMAR < 10 \mu m$ was calculated, based on either the data reported in the database (see also Section 3.5), personal communications from the authors of the original studies, or informed assumptions based on nearby observations as described in Albani et al. (2014)."

Comment 4287:17

The term of sediment accumulation rate is not applicable in the case of snow/ice being the archive matrix. **Response**

We changed to "sediment matrix", to be read in conjunction with the response above and the clarifications added to the text

Comment 4302:17

None of the cited references provide any error estimate for the reconstructed accumulation rate of the records used in this study. It follows, that the authors used the dating uncertainties to infer the errors of the accumulation rate which will yield far to optimistic error estimates.

It is inherently difficult to assess the uncertainty associated with reconstructed accumulation rates, especially if they are calculated from the age model of the ice core (which is the case for records here). In general the uncertainty is a combination of the errors of the vertical velocity (given by the dating error) as well as the uncertainty coming from the flow model used to correct for the thinning of the annual layers with increasing depth. Given the fact that the dating has been proven to be very accurate (especially in the Holocene), the uncertainty of the reconstructed accumulation rate is dominated by far by the error of the thinning function. This is, for example, very well illustrated in Kindler et al. (2014) for the last glacial, where the accumulation rate is reconstructed through $\delta 15$ N measurements. Thus only estimating the uncertainty through the dating error will in general yield to low error estimates.

I would advice the authors to contact the original authors of the reconstructions to provide an estimate of the

uncertainties of their thinning models. In any way, I would suspect the relative error of the accumulation rate to realistically be at least in the 10 % to 20 % range.

Response

The Referee raised an interesting point. We agree that our approach is prone to underestimate the uncertainties in the accumulation rates in general. On the other hand on the timescale of interest of this manuscript and for the ice cores records we present, we can expect significant less uncertainty in ice thinning for the Holocene sections than indicated by e.g. Kindler et al. Future extensions of the database to longer time scales, especially encompassing glacial stages, should address this problem more specifically as suggested. We clarify this in the text: "This is a reasonable approximation for the Holocene records from the ice cores presented here, but significantly larger uncertainties related to ice thinning models should be considered for deeper sections of ice cores and for glacial stages (Kindler et al., 2014)."

Comment 4314:22

The statement about changing deposition mechanisms as source of variability during the Holocene needs some further clarification. Unnerstad and Hansson (2001) have tackled this problem for the last glacial maximum, where due to the significant lower accumulation rate dry particle deposition has played a much larger role. However during the Holocene the accumulation rate is thought to be stable.

Response

We rephrase to be more specific, as suggested: "...changes in deposition mechanisms, which was suggested to be important on glacial/interglacial time scales but may be of minor relevance during the Holocene when accumulation rates are thought to be rather stable (Unnerstad and Hansson, 2001)."

Comment: Data Accessibility

Together with the manuscript the authors supply DMAR time series as well as the rebinned PSDs. Besides the missing information stated above it would be good if the quality labels that the authors assigned to the individual data sets would be provided in machine readable form as well. Additionally the authors state the possible application of their modeled global dust fluxes as input for other model studies. To facilitate that it would be good if the global fields of DMAR for the different time slices could be made accessible as downloads. **Response**

Good point. We included in the Supplement a table in text format with the indication of the geographical coordinates, the confidence level, and the availability of size data in the database for each of the 45 sites. In addition the modeled dust fields are available upon request.

Comment: Wrong Citations

There are two wrong citations in the text one of Ruth et al., 2003 and of Reid et al., 2003. Here the bibliography information used to generate the references is not correct.

Lately the bibliography export of the Whiley Journals has been acting up so it is probably a good idea to recheck all the used references.

Response

Thank you for pointing that out. We corrected the indicated references, and revised the References section.

Comment: Figures

In general the Figures provided with the manuscript are illustrative of S. Albani et al. 's argumentation, even though their layout is probably not final. In Figure 16 the labels of the individual datasets shown are often not visible because they overlap.

Response

I think that the figures were scaled to fit the pages in the online discussion format. The final figures should be reported in their original size, which is larger and more visible for Fig. 16 for instance.

Response to Referee #3 (F. Lambert)

Comment

The manuscript presents a collection of 122 dust flux Holocene time-series from various paleoclimatic archives in one database. Very importantly, the authors also include particle size data as well as estimates of the uncertainties for both the age and the dust flux data. Based on these uncertainties they select 43 records to represent global changes during the Holocene and use these to constrain dust simulations using the CESM. The work performed by Albani et al. is of great significance for the paleoclimatic dust community and provides a framework for expansion of the database. There are some issues that need to be addressed before it is ready for publication, though.

Above all, I would like to apologize for being late with my review. Very sorry about that.

Response

Dear Fabrice, thank you for your detailed comments. See our point-to-point response below.

Major Comments:

The major issue I have with this paper is the handling of the uncertainties. The uncertainty estimates are a very important part of this manuscript and the mathematics need to be described in much more details (can be in the summary). In each case (loess, marine, ice-core, peat) how are the final DMAR uncertainties actually calculated for data points (show the equations)? Are the errors normally distributed (discuss this for each archive)? If the errors are not normally distributed, how do the authors handle the uncertainty estimation and the error propagation (see the minor comments for more details about this)?

Response

The assumption we make is that the uncertainties are normally distributed. With reference to one specific comment (p. 4302, line 9), note that the non-Gaussian errors you mentioned actually refer to variables that we do not include in the calculations of the EC (Eolian Contribution) uncertainty (e.g. non-dust inputs etc.). Those are rather accounted for in the formulation of the categorical attribution of the confidence level. We think that it is reasonable to treat the errors as Gaussian in absence of better constraints, as well as the fact that this assumption is definitely of second order importance given the other uncertainties and available information. See the more detailed responses to specific comments below.

Minor Comments:

p. 4285, line 1: remove "formed by the accumulation of" **Response:** Fixed

p. 4285, lines 8-19: I wholeheartedly agree with this paragraph. How, concretely, did you address this problem, did you average the higher resolution record? Remember to check that the data are normally distributed before averaging, else use the median.

Response: We did not make any adjustments to the data in this respect. Note that we only have records where either the resolutions match or are very similar, as explained in the following lines. Some records were labeled as "low resolution" (see Table in the Supplement) because the SR could not be resolved during the Holocene. Specifically the issue only potentially applies in this compilation for medium level of confidence records i.e. for marine sediment records produced following the "operational approach", and it is actually the case only for V21-146. The resolution of both EC and SR for each record is visible from the ASCII tables in the Supplement. We added some explanation into the main text.

p. 4285, lines 23-26: I would add just one sentence here about how these problems are addressed. As it stands, the reader may think there's nothing to do about it.

Response: We added a sentence as suggested: "When there is indication of such occurrences, we either took focussing-corrected data in the former case, or considered only the undisturbed sections of the records in the latter case."

p. 4286, lines 2-4: Depends on your definition of "remote". I would argue that at a remote site you have no local input.

Response: Good point. We rephrased as follows: "In accumulation sites far from the major dust sources..."

p. 4286, lines 4-6: This sounds like there is a clear cut-off between short and long- range transport. Since size distribution is so fundamental in this paper, the background on dust sizes should be considerably expanded. **Response**: Excellent point. We have expanded the discussion on size distributions, as also suggested by Referee #1: we added Section 4.13 and Figure 18. Please refer to the revised version of the manuscript.

p. 4286, lines 23-24: The original reference for the nssCa is Röthlisberger 2002 GRL **Response**: We added the suggested reference

p. 4287, line 4: Narcisi reference is missing **Response**: We added the reference, thanks for pointing that out

p. 4288, line 4: Remove "linked to the carbon cycle". Either spend one or two sentences (with useful references) on the link or don't mention it at all. This whole sentence needs references, by the way.
Response: We removed the words about the carbon cycle. We added references in this paragraph and the following, as suggested.

p. 4288, line 9: . . ., as well as... **Response**: We fixed the typo, thanks for pointing that out

p. 4288, line 13: Reference about the Southern Ocean **Response**: We added a reference

p. 4290, line 3-7: Give some background and references on 232Th **Response**: That is explained and references are provided in the following lines

p. 4290, line 14: Add one original reference about end-member modeling. **Response**: Reference added

p. 4295, lines 24-25: The Gaussian distribution of the errors is a fundamental condition for the follow-up steps in this paper and cannot just be assumed. See my comment for p. 4302, line 9.
Response: Please see the response below to the comment indicated by the referee.

p. 4298, line 22: Narcisi reference is missing.Response: We added the reference, thanks for pointing that out

p. 4298, line 24: Gabrielli reference is missing (better check them all). **Response**: We added the reference, thanks for pointing that out

p. 4298, lines 26: Talking about EC when meaning dust in ice is a bit confusing as everything in an ice-core, including the ice, falls from the sky. Why not just talk about particle concentrations?
Response: We think our terminology is clear enough in this context, as we are obviously referring to dust as stated in Section 2, which reads: "dust (or eolian – the two terms will be used equivalently throughout the text)".

p. 4298, lines 26-27: Un-calibrated laser data (in Volt or P/ml) is useless for determining the dust flux and not a "critical" uncertainty. If size distribution is present, though, then a conversion of P/ml to mass concentration is possible.

Response: We actually refer to a calibration of the size i.e. against coulter counter measurements (Ruth et al.,

2003). Assumptions on the shape and optical properties of the particles inherent in the conversion of the laser's reading into volume size distributions can introduce significant biases (e.g. Reid et al., 2003). While for instance calibration of laser counters with latex spheres is common in the procedures used to determine the size distributions in the marine community, they also determine the MARs independently, so that possible biases are of second order importance. On the other hand for an ice core where both the size distribution and the EC are determined in such a way, errors can be substantial. We clarify this in the text: "Un-calibrated (for the size) laser counters give unreliable results, as both the size distributions and the EC may be significantly affected, which we consider a critical uncertainty".

p. 4298, line 28 - p. 4299, line 1: Which data in the Ruth et al papers signifies that Ca is a better proxy for dust in Greenland than Antarctica? The higher Greenland Ca:dust ratios in the first Ruth et al paper are doubtful considering the difficulties with the Laser calibration especially in that early stage. Steffensen et al., 1997 for example published similar Greenland Ca:dust ratios as found in Ruth et al., 2008 in Antarctica.
Response: the uncertainty refers to the proportions of crustal versus nss-Ca in the two cases, with a sea salt deposition one order of magnitude higher than dust in Antarctica, but much lower in Greenland. We clarified this in the text.

p. 4299, line 3: The "significant uncertainty" is confusing here. Is this a general statement or did you mean substantial as a flag?

Response: Substantial. We clarify that.

p. 4301, line 14: This formula would be correct if you added the two ages on both sides. A linear interpolation, however, is of the form Asample = a*A1+b*A2 with a and b between 0 and 1, depending on how close you are to one or the other endpoint. The error for the sample is then Esample = sqrt(a2 E12 + b2 E22) (With carbon dating we can assume no covariance between the two errors). You can get the a and b values from the distance of each depth horizon from the two dated horizons.

Response: Good point. Note however that this assumption is definitely of second order importance given the available information, and when considered in the general context. As we explain in Section 3, in this work the uncertainty in the records is composed by two elements: (a) the categorical attribution of confidence, and (b) quantification of the uncertainties related assumptions and to analytical uncertainties. The largest uncertainties are likely contained in (a). The quantification in (b) is meant to be a first order approximation of the quantifiable uncertainties, which are themselves subject to approximation and subjective evaluation to some extent (see also response below). We clarify this in the text: "Note that a large part of the actual uncertainties associated with each record are related to what we include in the attribution of the confidence level, and that the estimates provided for the quantifiable uncertainty constitute a first order approximation".

Note that this point would be of more relevance if we focused on re-deriving the original age models, which was beyond the scopes of our work at this stage.

p. 4301, line 22: I don't understand this. I see that 6.8% is 10% of one standard deviation, but I don't see the reasoning behind that value.

Response: It is an arbitrary choice, as stated in the text, which we consider to be reasonable.

p. 4302, line 9: This is where we run into problems with the uncertainties. First, the equation is ϵ MAR / μ MAR = sqrt(...). Second, how were ϵ SBMAR , μ SBMAR , ϵ EC , and μ EC calculated in various datasets (see my Major Comment)? I don't know about SBMAR, but the uncertainty in EC will be a sum of many errors, some of which are normally distributed (e.g. analytical errors, bioturbation?) and some of which are not (e.g. tephras and local source contribution in ice-cores, volcanic and lithogenic input in marine sediments, etc.), which means that ϵ EC is not a standard deviation in the Gaussian sense. You could argue that they are small enough that the Central Limit Theorem is not violated. However, all of the non-Gaussian errors I can think of are positive. Finally, what does μ represent, what exactly did you average? It should be the variable whose standard deviation is ϵ .

errors you mentioned actually refer to variables that we do not include in the calculations of the EC uncertainty (e.g. non-dust inputs etc.). Those are rather accounted for in the formulation of the categorical attribution of the confidence level.

The error propagation formula for the case of multiplication is applied in Equation 2. The ratio epsilon/mu represent the relative error, i.e. the error divided by the absolute value it refers to. The details of how SBMAR and EC were derived, and what the explanation is for the estimation of the errors, are reported for each record in the Supplement – see the individual "descriptive sheets". The cases we encountered are discussed in Section 2 for specific types of archives, as well as in the Methods Section 3.3.

As described in the text, some of these errors indeed are associated to replicate measurements of the same sample, like we report for Antarctic ice cores' EC associated to Coulter Counter measurements for instance, or as is the case thorium measurements. In those cases mu represents the average of the measurements, and more in general it is the absolute value. On the other hand in other cases the uncertainty is arbitrarily assigned based on an expert informed guess, given the lack on information in the literature on specific estimates. We think that it is reasonable to treat the errors as Gaussian in absence of better constraints, also considering the fact that this assumption is definitely of second order importance given the available information. We clarified this in the text, by replacing "average" with "absolute values".

p. 4302, line 20: Why 5%? Explain or provide reference.

Response: as already explained, the uncertainty here is arbitrarily assigned based on an expert informed guess. We reiterate this in the text.

p. 4303, line 16: What's the spread of the analytical uncertainty in those records where it is available? **Response**: it is difficult to assign an analytical uncertainty to size distributions. Note that this is of second order importance compared to the possible biases and differences among different measurement methods (e.g. Reid et al., 2003 or Mahowald et al., 2014). We report objective metrics of the re-binning procedure, though – see supplement.

p. 4303, lines 17-20: How did you get the 26%? Based on the Steffensen data I get a Ca:dust ratio of around 1:100 also for warm Holocene times. As already mentioned, the Ruth et al., 2002 data for dust is unreliable due to the difficulty in calibrating the early lasers. Why not use the stdev of the calcium:dust ratio in the Steffensen data as a proxy-uncertainty and combine it with the analytical uncertainty instead of the arbitrary 20%?

Response: We referred to Steffensen (1997) and Ruth et al. (2002) for the Ca/dust relation, and took the average for both "warm" (0.26) and "cold" (0.095) periods, based on GISP2 d18O record. We assumed "warm" if d18O>-37 per mil, and cold if d18O<-40, and used linear interpolation among those two values for the range in between. I quote Ruth et al. (2002): "A (Ca2+)/(insoluble dust) mass ratio of 0.29 was found for Holocene and 0.11 for LGM" – and Steffensen (1997): "the mass ratio of calcium and dust during the LGM was 0.08 compared to 0.23 during the Holocene". See the explanations in the relative descriptive sheet.

As for the uncertainty, we follow up based on your suggestion, which sounds like a good idea actually. Because we refer to two different studies as explained above, we consider instead a calcium/dust uncertainty of ~11.5%, i.e. (0.29-0.26)/0.26. Using equation 1 to combine that one and the analytical uncertainty would give the total proxy uncertainty. To have a total proxy uncertainty of ~20% as we first suggested, it would imply an analytical uncertainty of ~18%, which is quite reasonable for GISP2 Holocene samples, with an average Ca2+ concentration ~8 µg/kg. For instance Ruth et al. 2008 suggest that, with reference to soluble calcium from ion chromatography, samples with concentrations ~20 µg/kg have analytical reproducibility better than 10%, whereas lower concentration samples ~1 µg/kg would bear uncertainties around 100%.

p. 4307: line 2: MAR is spatially more or less log-normally distributed. Averaging over an area will bias the data towards high values! Use the median in these cases.

Response: We specify that we do this "For each record". The anomalies to the reference period are then averaged within each macro-region, as explained in the following lines. Averaging is therefore appropriate.

p. 4307, line 3: Are you talking about spatial gaps or temporal gaps here? In both cases you have to take into account that dust MAR is spatially and temporally close to log-normally distributed. You should therefore use linear interpolation on the logarithms of the MAR data and then transform back. **Response**: Temporal gaps. See the previous response.

p. 4312, line 27: Maybe include Kukla in the list of citations here? **Response**: we added a reference to Kukla and An (1989). Thanks for the suggestion.

p. 4320, lines 10-23: Are the records flagged with low confidence going to be included in the database? So far, only the 43 records with high and medium confidence are included. **Response**: No records with low confidence level will be included in the database, as explained in Section 3.1.

p. 4320, lines 13-14: The ASCII files are probably the best way to make it readable for the largest amount of people. In addition to those, I think a NetCDF file would be very handy so one can download all the data in only one file.

Response: The NetCDF format is not the most suitable one for organizing such kind of data. It is already possible to download all the data conveniently in one file (zip archive).

p. 4323, lines 14-16: I would expect the sum of all size bins to result in the Dust MAR value for that sample (second column), but this doesn't seem to be the case. What value is given in each size bin? Also, there seems to be a problem with the EDC data; three of the first 4 columns are missing and some samples seem to have their values shifted to small bins. There are some negative values in the Zagoskin data (should probably be zero). **Response**: The negative values in fact are rounding errors, as suggested by the very small values. We fixed that. Thanks for pointing that out. We re-processed the data in order to explicitly conserve the information on the size range, as suggested by Referee #2. The numbers in each of the new size bins represent the percentage contribution to the mass/volume in the total size range, so the sum of the defined bins should be 100. We add text to the Appendix to clarify this.

As for EDC data, they are fine. There is no mistaken shift in the small bins: the lower limit of the size range is subject to very small variations in relation to the calibration setup of the Coulter Counter.

p. 4323, lines 22-24: Could you explain the figures (in the folder /Database/Size_Description) a bit more here? I'm guessing red is the spline and black and green are the original data? How were the samples used for the figures cho- sen? For EDC and Vostok it looks like the size distribution was truncated at 5 um (and maybe also at the lower tail). Maybe Barbara can confirm if this is the case? If so, the model may be improved by extrapolating the spline (both EDC and Vostok seem to be overestimated by the spline).

Response: We added some description in the Appendix.

As for EDC and Vostok, as explained in the specific "descriptive sheets", the size distributions were truncated at 5 um, which we consider as the limit where the number of the particles is significantly different than a blank for those two records (e.g. Delmonte et al., 2013).

p. 4324, lines 1-5: I don't think the MNB is much used outside the modeling community. I suggest to add a few sentences that explain this metric and how to read it.

Response:

We clarified this: "In this context, the MNB is a metric of the average over- or under-estimation of the "coarseness" of the re-binned size distributions compared to the original observations".

Figure 4: Units on lower panel y-axis

Response: It's unitless, we now display that.

Figure 5: blue = low confidence? I'm not sure the green dots add much information here. Either there's only one dot that is probably from the same record that is plotted, or they cover the whole range and thus do not really confirm anything. I suggest to either discuss the comparison with DIRTMAP3 data in page 4308 or to remove these from the plot.

Response: "blue = medium confidence", we fixed the legend, thanks for pointing that out.

The comparison to DIRTMAP3 highlighted by the green dots in Figure 5 shows the density of information within each region, and the span of the temporal variability. No confirmation is sought there. We present this piece of information to give an overview of our data in the same context with the reference work for paleodust, while highlighting the extra information we provide in terms of temporal resolution.

Other comments: I have a general question about the 230Th method: Is the difference in sea-level between LGM and Holocene taken into account? This is obviously not of any concern for this paper, but may be an issue for the extension of the method to the LGM.

Response: Good question. This aspect is usually accounted for in the original studies.

1 Twelve thousand years of dust: the Holocene global dust

2 cycle constrained by natural archives

3

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19

20 Abstract

Mineral dust plays an important role in the climate system by interacting with radiation, clouds, and biogeochemical cycles. In addition, natural archives show that the dust cycle experienced variability in the past in response to global and local climate change. The compilation of the DIRTMAP paleodust datasets in the last two decades provided a target for paleoclimate models that include the dust cycle, following a time slice approach. We propose an innovative framework to organize a paleodust dataset that moves on from the positive experience of DIRTMAP and takes into account new scientific challenges, by providing a Cornell University 3/5/15 10:01 AM Deleted: 12

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concise and accessible dataset of temporally resolved records of dust mass accumulation rates 1 2 and particle grain-size distributions. We consider data from ice cores, marine sediments, 3 loess/paleosol sequences, lake sediments, and peat bogs for this compilation, with a temporal 4 focus on the Holocene period. This global compilation allows investigation of the potential, 5 uncertainties and confidence level of dust mass accumulation rates reconstructions, and highlights the importance of dust particle size information for accurate and quantitative 6 7 reconstructions of the dust cycle. After applying criteria that help to establish that the data 8 considered represent changes in dust deposition, 45 paleodust records have been identified, 9 with the highest density of dust deposition data occurring in the North Atlantic region. 10 Although the temporal evolution of dust in the North Atlantic appears consistent across several cores and suggest that minimum dust fluxes are likely observed during the Early to 11 12 mid-Holocene period (6,000-8,000 years ago), the magnitude of dust fluxes in these 13 observations is not fully consistent, suggesting that more work needs to be done to synthesize 14 datasets for the Holocene. Based on the data compilation, we used the Community Earth 15 System Model to estimate the mass balance and variability of the global dust cycle during the 16 Holocene, with dust load ranging from 17,2 to 20,8 Tg between 2,000 and 10,000 years ago,

17 and a minimum in the Early to Mid-Holocene (6,000-8,000 years ago).

18

19 **1** Introduction

20 Paleoclimate records from natural archives have laid foundations for understanding the 21 variability of the Earth's climate system over different time scales. Paleoclimate proxies shed 22 light on past environmental conditions such as the composition of the atmosphere, global ice 23 volume, sea level, and surface temperatures (Bradley, 1999). Paleodust reconstructions paired 24 with other proxies showed the response of the climate system to orbitally induced forcing, 25 including feedback mechanisms. Dust feedbacks on the climate system include scattering and 26 absorption of solar radiation and indirect effects on clouds and the global carbon cycle (e.g. 27 Boucher et al., 2013; Martin, 1990).

The story told by paleodust archives suggests that increased aridity (An et al., 1991; Liu, 1985; Liu et al., 1998) and wind gustiness (McGee et al., 2010; Muhs et al., 2013) enhanced the dust cycle during cold periods over glacial-interglacial time scales, with additional mechanisms introducing characteristic geographic patterns and/or imprinting the archives with characteristic signals in different geographical settings. These mechanisms include Cornell University 3/5/15 10:01 AN **Deleted:** 43

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1 increased sediment availability by glacial erosion (Delmonte et al., 2010a; Petit et al., 1999),

2 reorganization of the atmospheric circulation between mid and high latitudes (Fuhrer et al.,
3 1999; Lambert et al., 2008; Mayewski et al., 1997, 2014), shifts in the Inter-Tropical

4 Convergence Zone (ITCZ) (McGee et al., 2007; Rea, 1994), changes in the monsoonal 5 variability (Clemens and Prell, 1990; Hovan et al., 1991; Tiedemann et al., 1994), and

6 regional drying (Lu et al. 2010).

7 The growing number of paleodust archives and the inclusion of the dust cycle in climate 8 models has promoted synthesis efforts in the compilation of global dust datasets (Mahowald 9 et al., 1999). The Dust Indicators and Records from Terrestrial and MArine 10 Palaeoenvironments (DIRTMAP) Project (Kohfeld and Harrison, 2001) formalized the 11 compilation of Dust Mass Accumulation Rates (dust MAR, or DMAR) from marine and ice 12 cores, later complemented by terrestrial sedimentary records (Derbyshire, 2003). This project 13 followed a time slice approach, providing reference values of DMARs for the Last Glacial 14 Maximum (LGM) and Late Holocene / modern data, including sediment traps. DMAR is the 15 fundamental measurement necessary to cross-correlate variability among dust archives and 16 sites. Without it, only the relative timing and amplitude of individual records can be studied. 17 In combination with global climate models, DMAR datasets enable quantitative 18 reconstructions of the global dust cycle. The DIRTMAP compilation showed a globally 19 averaged glacial/interglacial ratio of ~2.5 in dust deposition. Subsequent work expanded upon 20 the initial compilation (DIRTMAP2: Tegen et al., 2002), and the most recent version of the 21 database (DIRTMAP3: Maher et al., 2010) also contains an extensive repository of additional 22 metadata from the original publications. The DIRTMAP datasets have proven to be an 23 invaluable tool for paleoclimate research and model-data inter-comparison.

24 The full definition of the global dust cycle in terms of DMAR is unavoidably linked to the 25 dust grain size distributions that characterize the mass balance and its spatial evolution. The more advanced dust models define a model particle size range and distribution, which would 26 27 require (although this has been often neglected) explicitly considering the size range of dust 28 found in the dust deposition data in model-observation inter-comparisons. This aspect was 29 initially taken into account for terrestrial sediments in Mahowald et al. (2006) to match the 30 specific model size range $(0.1-10 \ \mu m)$, and recently extended by Albani et al. (2014). Still the necessity of more extensive grain size information from dust data has been emphasized by 31 32 Maher et al. (2010), as well as by other review papers on dust (e.g. Formenti et al., 2011;

1 Mahowald et al., 2014). Coherent information on grain size is missing in DIRTMAP3 (Maher

2 et al., 2010), because of the difficulty of making a synthesis from measurements produced by

3 a variety of particle-size measurement techniques often yielding quite different results

4 (Mahowald et al., 2014; Reid, 2003).

5 A time slice approach is often used by the paleoclimate modelling community to target key periods in climate history, such as the Last Glacial Maximum ~21,000 years Before Present 6 7 (LGM: 21 ka BP), or the Mid-Holocene (MH: 6 ka BP), in the framework of the Paleoclimate 8 Modelling Inter-comparison Project (PMIP: Joussaume and Taylor, 2000). Continuing 9 improvement in the performance of large-scale supercomputers is opening up doors to 10 performing transient simulations on paleoclimate time scales, both to intermediate complexity 11 (Bauer and Ganopolski, 2014) and more complex Earth System Models (ESMs) (Liu et al., 12 2009). PMIP3 called for additional key transient experiments to study abrupt climate change, 13 with the implication that at the same time target observational datasets with the necessary 14 temporal continuity and resolution are needed (Otto-Bliesner et al., 2009).

15 We propose an innovative framework to organize a paleodust dataset that moves on from the 16 positive experience of DIRTMAP and takes into account new scientific challenges outlined 17 above, by providing a synthesized and accessible dataset of temporally resolved records of 18 dust MARs and size distributions. We aim to provide a database that is a concise and 19 accessible compilation of time series, including age (with uncertainty), dust MAR (with 20 uncertainty), and dust particle size distribution (where available), standardized by the use of a 21 common binning scheme, and complemented by a categorical attribution of confidence based 22 on general consensus. Besides the basic information mentioned above, we also report the 23 ancillary information necessary to re-derive the dust MARs time series, i.e. the detailed 24 depths and the relevant dust variables. Inspired by DIRTMAP, our new compilation considers 25 DMARs as the key variable for a coherent study of paleodust archives. The elements of 26 innovation that we introduce here (size distributions, temporal resolution, and attribution of 27 confidence level) however constitute a leap forward into a new generation dust database.

We focus on dust variability during the Holocene, with emphasis on the MH as a key PMIP scenario and also in relation to the large variability that affected the present largest dust source in the world, North Africa, with the termination of the African Humid Period (AHP) (deMenocal et al., 2000; McGee et al., 2013). For this reason we only selected paleodust records encompassing the MH with some degree of temporal resolution (see Sect. 3),

- 1 although we show in the paper the time series from the LGM to provide reference to other key
- 2 climate conditions and to place in a fuller context with respect to the DIRTMAP compilation.
- 3 The developed framework is suitable for a more extensive compilation.

4 We acknowledge that there is a richness of information intrinsic in each sedimentary record 5 (i.e. as in the original studies) that is not necessarily fully captured by the synthesized information we report, despite our efforts to be as complete as possible: simplification is 6 7 inherent in a synthesis. For the sake of accessibility we refrain from reporting extensive 8 information that cannot be coherently organized. We therefore provide a brief summary, and 9 refer to the relevant literature for detailed description of specific records (Supplementary 10 material). In addition, because our purpose is to provide a quantitative constraint on the dust 11 cycle, we only considered sedimentary records that allow the derivation of meaningful dust 12 MARs with the information we could access. Many more studies focused on dust and provide 13 important, good quality information, but did not allow a time-resolved estimate of dust MAR. 14 We refer to these studies when appropriate, as they provide further context to ensure our 15 interpretations.

Finally, we use the Community Earth System Model (CESM) in combination with the DMAR
and size data (Albani et al., 2014; Mahowald et al., 2006) from the compilation to estimate the
mass balance of the global dust cycle and its variability during the Holocene.

Section 2 gives an overview of the kind of natural archives initially considered for this compilation, while in Sect. 3 we explain our methodological approach to select and organize the records. In Sect. 4 we present the database and model-based reconstructions, and discuss its emerging properties in relation to the climate features in different spatial domains. We summarize our work in Sect. 5.

24

25 2 Paleodust archives

Natural archives that preserve dust sediments have different characteristics in terms of: geographical settings and spatial distributions around the globe; the accuracy of the age models and temporal resolution; the ability to isolate eolian dust from other depositional contributions. Each type of paleodust archive has its own strengths and limitations, and it is only by considering high quality records of all types (from land, ice, and ocean archives) that we can hope to build a consistent reconstruction of the global dust cycle. We only include

1 paleodust records that allow estimation of dust MARs with relevance for medium/large scale

2 dust export.

Natural archives preserve eolian dust within a sedimentary matrix. The essential elements for
a paleodust record are the possibility of establishing a reliable chronology, the estimation of
the sedimentation rates, and the isolation of the eolian component (Fig. 1). Throughout the
paper we use the term "sediment" in a broad sense that encompasses ice as well as other
sediments in a strict sense.

One of the key elements in the production of a paleodust record is the possibility of 8 9 establishing a depth-age relation. Typically the starting point for this procedure is the 10 attribution of age to a series of specific depth layers along the profile, based on numerical 11 dating or stratigraphic correlations. Numerical dating can be based on counting of annual layers, radionuclide decays (e.g. ¹⁴C), or exposure to radiation (e.g. Thermo-Luminescence 12 13 (TL) / Optically Stimulated Luminescence (OSL)) (Brauer et al., 2014). Stratigraphic 14 correlations either exploit stratigraphic markers such as known volcanic eruptions and spikes 15 in tracers of the atmospheric thermonuclear test explosions, or are attributed by wigglematching an age-carrier profile from the study site (e.g. δ^{18} O of foraminifera in marine 16 17 sediment cores, methane concentration in ice cores) with a reference record of global 18 signatures such as global ice volume (e.g. Martinson et al., 1987), or the variations in 19 atmospheric methane concentrations (e.g. Loulergue et al., 2008).

Sediment chronologies can be established based on the initial age-depth relations identified along a profile. With "chronology" we identify a continuous function that provides a unique attribution of the depth-age relation along the entire profile, based on some kind of age model. Age models can vary from simple linear sedimentation models, to complex Bayesian models (Brauer et al., 2014).

A general expression for dust (or eolian – the two terms will be used equivalently throughout
the text) MARs is the following: DMAR = SBMAR * EC, where SBMAR is the Sediment
Bulk Mass Accumulation Rate and EC is Eolian Contribution.

The estimation of SBMAR relies on a couple of main approaches. The first one is based on estimating SBMARs between dated horizons as the product of sedimentation rates and dry bulk densities: SBMAR = SR * DBD. Either a Linear Sedimentation Rate (LSR) is derived between dated layers, or more complex age models are applied, resulting in diverse SR profiles. The other approach is specific for the marine sediments realm, and it is largely (other

1 than for decay-correction) independent from the underlying age model: it is based on the 2 assumption that the rapid scavenging of ²³⁰Th produced in the water column by decay of 3 dissolved uranium results in its flux to the seafloor being equal/close to its known rate of 4 production. Measurements of ²³⁰Th in marine sediments therefore allow us to estimate 5 instantaneous SBMARs that are independent from LSRs (François et al., 2004).

6 Because eolian DMAR is the product of at least two factors (SBMAR and EC), the sampling 7 (depth) resolution at which the two of them are available will determine the DMAR 8 resolution, and in some cores the resolutions may coincide. Sometimes a constant LSR is 9 assumed between dated depth layers whereas stratigraphic samples are analysed at higher 10 resolution and an estimated age is assigned based on the age model (Fig. 2). At the time scale 11 of interest, it should be noted that deviations from the ideal pairing of EC and SBMAR 12 measurements along a profile might be considered acceptable if the resolutions are not too 13 different. On the other hand, if one variable (typically EC) has a much higher resolution than 14 the other, then its high resolution is not informative with respect to their product (DMAR), 15 and misinterpretations could arise. In those cases the lower resolution variable should be used 16 to provide the pace of the record's resolution. We did not make any adjustments to the data in this respect; note that we only have records where either the resolutions match or they are 17 18 very similar (see Supplement).

19 An additional aspect to consider when dealing with dust MARs is the relationship between the 20 dust Deposition Flux (DF) and the dust MAR i.e. to what extent the measured DMAR is representative (in a quantitative way) of the dust deposition, which is of primary interest: 21 22 ideally DMAR = DF. Deviations from this ideal relation occur, for instance, when sediment 23 redistribution disturbs the ocean sediments (François et al., 2004), or when erosion leaves hiatuses in loess/paleosol sequences (Stevens et al., 2007). When there is an indication of 24 25 such occurrences, we either took focussing-corrected data in the former case, or considered 26 only the undisturbed sections of the records in the latter case.

The other fundamental piece of information is the size distribution of dust, which is tightly coupled to the DMAR in determining the magnitude (or mass balance) of the dust cycle (Albani et al., 2014; Mahowald et al., 2014; Schulz et al., 1998; Lu et al., 1999). In addition, size data is a necessary piece of information to determine the provenance of dust. In accumulation sites far from the major dust sources, size distribution allows (together with geochemical and mineralogical data) the identification of local versus remote inputs (Albani

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1 et al., 2012a; Delmonte et al., 2010b). In terrestrial sites proximal to the source areas it is

2 necessary to evaluate the amount of dust actually available for long-range transport

3 (Mahowald et al., 2006; Muhs et al., 2013; Roberts et al., 2003).

4 We next analyse the main characteristics of the different kinds of paleodust records 5 considered for this compilation: ice cores, marine sediments, loess/paleosol sequences, lake 6 sediments and peat bogs.

7 2.1 Ice cores

8 Ice cores constitute a natural sampler of past atmospheric composition, including greenhouse 9 gases and aerosols. Isolation of the eolian component from the ice matrix is rather 10 straightforward – it is usually obtained by melting the ice at room temperature (Delmonte et 11 al., 2004), although sublimation of the ice is another option (Iizuka et al., 2013) – so that the 12 ice allows the most pristine preservation of the locally deposited atmospheric aerosol.

13 The presence of perennial ice limits the geographical coverage of ice core records worldwide, 14 and the recovery of long dust stratigraphies is limited to the high latitudes and a few alpine 15 glaciers in the low and mid latitudes. Often the EC is a direct measure of the insoluble dust 16 concentration and size distribution in the ice samples, using either a Coulter Counter (Delmonte et al., 2004) or a laser diffraction particle counter (Lambert et al., 2008). 17 18 Alternatively a geochemical dust proxy can be used (e.g. McConnell et al., 2007), and the 19 most common approach considers non-sea salt calcium (Röthlisberger et al., 2002; Fischer et 20 al., 2007). Despite the fact that the dust-calcium relation should be taken with caution under 21 certain circumstances (Ruth et al., 2002, 2008), this approach has successfully been used to 22 produce dust records in Greenland (e.g. Fuhrer et al., 1999; Mayewski et al., 1997) and 23 Antarctica (Lambert et al., 2012; Schüpbach et al., 2013).

Since in most cases both dust (insoluble) and calcium records were produced at the same location, we focus on insoluble particle records, which also include dust size distributions. Possible non-dust contributions include volcanic tephra, which are usually identifiable and excluded from the records (e.g. Narcisi et al., 2012). For Greenland there is only one record spanning the Holocene, GISP2, for which we consider calcium as a proxy for dust (Mayewski et al., 1997).

For the estimation of SBMAR, post-depositional changes may potentially affect snow/ice accumulation rates through surface redistribution or sublimation. In the polar ice sheets

1 plateaus these effects are probably negligible on domes where ice cores are usually drilled

2 (Frezzotti et al., 2007), so that dust DMAR = DF.

3 Polar ice cores' age models are in continuous evolution and they benefit from the growing 4 number of deep ice cores. The striking feature is the absolute counting of annual layers in 5 Greenland ice cores (Vinther et al., 2006), which in combination with several ice and 6 stratigraphic markers (e.g. methane spikes, volcanic signals) allows establishing consistent 7 chronologies for both Greenland and Antarctic ice cores. In this work we use the most recent 8 AICC2012 chronology for Antarctic ice cores (Veres et al., 2013). Because of the high 9 sediment matrix accumulation rates compared to other natural archives, polar ice cores 10 usually provide the highest resolution dust records. Dust concentration records are also 11 available from alpine glaciers (e.g. Thompson et al., 1995, 1997). While it is possible to 12 derive estimates of dust MARs on the glacial/interglacial time scale (Kohfeld and Harrison, 2001), it is problematic to calculate DMAR time series. This is because there are no reliable 13 14 age models due to the difficulty in establishing adequate accumulation stratigraphies in such 15 environments.

16 With a few exceptions from sites on the edges of the ice sheets both in Greenland (Renland:

17 Hansson, 1994) and Antarctica (e.g. TALDICE: Albani et al., 2012a; Delmonte et al., 2013),

18 polar ice cores are thought to archive almost exclusively dust from remote source areas (Bory

19 et al., 2003; Delmonte et al., 2010b), and to be representative of the magnitude and variability

20 of the dust cycle at least over the high latitudes on both hemispheres (Mahowald et al., 2011).

21 2.2 Marine sediments

22 With the oceans covering two thirds of the Earth's surface marine sediment cores represent 23 key paleoclimate archives, recording among other things global land ice volumes, ocean 24 productivity and the main characteristics of the ocean deep circulation, (e.g. Bradley, 1999). 25 Dust particles deposited to the ocean's surface attach to other suspended particles and get 26 scavenged throughout the water column, determining the accumulation of eolian material in 27 pelagic sediments, (Bory and Newton, 2000). Despite the complexity and uncertainties in the 28 dynamics of particle sedimentation throughout the water column (e.g. Bory and Newton, 2000; De La Rocha et al., 2008), as well as their potential advection downstream (Siegel and 29 30 Deuser, 1997; Han et al., 2008), we can reasonably make the approximation that dust 31 DF(surface) = DF(benthic). This is valid in most regions (Siegel and Armstrong, 2002;

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Kohfeld and Tegen, 2007), with the notable exception of the Southern Ocean, (Kohfeld and
 Harrison, 2001).

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The pelagic environment is characterized by low deposition rates, so that most marine records naturally have a lower temporal resolution than ice cores. Chronologies for marine sediment cores are often derived by stratigraphic correlation of δ^{18} O records of benthic or pelagic foraminifera (representative of a combination of global ice volume and temperature) with reference stacks such as SPECMAP (Imbrie et al., 1984; Martinson et al., 1987) or LR04 (Lisiecki and Raymo, 2005).

9 In many studies, which is especially relevant for the Holocene, additional constraints for the 10 age models are given by radiocarbon-dating foraminifera (e.g. Anderson et al., 2006; McGee 11 et al., 2013) or tephras (Nagashima et al., 2007). The age-depth relation is usually assigned by 12 linear interpolation between dated layers. Chronologies only based on stratigraphic correlation of δ^{18} O records are inherently affected by a significant degree of uncertainty for 13 the Holocene, because the youngest tie-points in δ^{18} O stacks can be considered the last glacial 14 15 maximum (18 ka BP) and the Marine Isotopic Stage (MIS) boundary MIS1/2 (14 ka BP) 16 (Lisiecki and Raymo, 2005). Often, in the absence of absolute ages, the assumption is made 17 that the surface sediment age is 0 ka BP, although the surface sediments may be disturbed or 18 partially lost during the core recovery.

19 Two main strategies are used to derive dust records from marine cores. In the first, more 20 traditional "operational" approach SBMAR = LSR * DBD, with LSR calculated from the age 21 model and DBD measured or estimated. EC is determined by isolating the lithogenic fraction 22 from the sediment matrix by subsequent removal of the organic component, carbonates, and 23 biogenic opal by thermal/chemical treatments (Rea and Janecek, 1981). In this approach the 24 basic assumption is that the entire lithogenic fraction is eolian in origin. Corrections for 25 volcanic contributions were attempted by visual inspection (Hovan et al., 1991) or by the use of geochemical tracers (Olivarez et al., 1991), which could also help to distinguish fluvial 26 27 versus eolian inputs (Box et al., 2011). Other spurious lithogenic inputs may include material 28 from turbidite currents, hemipelagic sediments, or ice-rafted debris (e.g. Rea and Hovan, 29 1995). Additionally, sediment redistribution may alter the depositional stratigraphy biasing 30 the true sedimentation rates (François et al., 2004), which is usually not accounted for in 31 studies following this kind of approach. Here we exclude sites known (or very likely) to be 32 significantly affected by sediment redistribution (e.g. nepheloid layers: Kohfeld and Harrison,

2001), ice-rafted debris (Kohfeld and Harrison, 2001), and those close to the continental
 margins (e.g. Serno et al., 2014).

The other strategy consists in deriving SBMAR from ²³⁰Th profiling (François et al., 2004). 3 Briefly, 230 Th (half-life = 75,690 years) is produced uniformly throughout the ocean by 4 radioactive decay of dissolved ²³⁴U. Due to its high particle reactivity, ²³⁰Th is efficiently 5 scavenged by particulate matter and has a short residence time in the ocean (< 30 years) 6 (Bacon and Anderson, 1982). The rain rate of scavenged ²³⁰Th to the sediments is therefore 7 equal to its known rate of production in the overlying water column (Henderson et al., 1999). 8 9 SBMARs are calculated by dividing the production rate of ²³⁰Th in the water column by concentrations of scavenged ²³⁰Th in the sediment (Bacon, 1984; François et al., 2004). 10 At sites potentially influenced by sediment redistribution, the ²³⁰Th profiling method is

11 12 probably the more reliable approach for the determination of SBMAR, as it accounts for sediment focusing (Anderson et al., 2008; François et al., 2004). If it can be assumed that the 13 lithogenic fraction is of eolian origin, EC can be derived from the ²³²Th concentration in the 14 sediment of a dust proxy (²³²Th). As ²³²Th concentrations in dust are generally more than an 15 order of magnitude higher than in most volcanic materials, ²³²Th levels closely track 16 continental inputs and are insensitive to volcanic inputs. In addition, ²³²Th offers the 17 advantage compared to other dust proxies, that its concentration in global dust sources is 18 19 relatively invariable and close to the upper continental crust concentration (McGee et al., 20 2007). If non-eolian contributions (such as volcanic) are present, multi-proxy approaches (using REE, ⁴He) can provide a means to isolate the eolian fraction (Serno et al., 2014). On 21 22 continental margin settings high sedimentation rates are related to the presence of fluvial 23 inputs, which can be isolated from the eolian component by use of grain size end-member 24 modelling (McGee et al., 2013; Weltje, 1997).

Bioturbation i.e. surface sediment mixing by the benthic fauna is a common unconstrained feature of marine sediments, that acts as a smoothing filter on the sedimentary stratigraphy, including ages and other profiles interest, with a typical vertical smoothing scale of 8-10 cm. A few studies evaluated the potential effects of bioturbation of their records, although they do not correct their profiles (François et al., 1990; McGee et al., 2013), based on a simple deconvolution linear model (Bard et al., 1987).

1 2.3 Loess/paleosol sequences

The possibility of reconstructing the global dust cycle requires observations distributed 2 3 geographically to constrain different regions, but also encompassing the evolution of dust 4 spread from the source areas to the areas downwind and to remote regions. Terrestrial 5 sediment records are therefore necessary to constrain the location and magnitude of past 6 source of dust. Loess can be defined as terrestrial eolian sediments, composed predominantly 7 of silt-size particles, formed by the accumulation of wind-blown dust (Pye, 1995; Liu, 1985), 8 covering vast regions (~10%) of the land masses (e.g. Derbyshire et al., 1995; Rousseau et al., 9 2011). The formation of loess deposits is often associated with the proximity of major dust 10 sources, the availability of fine-grained erodible sediments and adequate winds, and a suitable 11 accumulation site (Pye, 1995; Liu, 1985). This requires that a complex deposition-erosion 12 balance determines the actual rate of accumulation at a site and the alternation of 13 accumulation / weathering phases depending on the dominant environmental conditions 14 (Kemp, 2001; Muhs et al., 2003a). Loess/paleosol records (or soil profiles) spanning the Late 15 Ouaternary have shown to be important proxies and dust archives, both on glacial-interglacial 16 (e.g. Kohfeld and Harrison, 2003; Muhs et al., 2008; Lu and Sun, 2000; Liu et al., 1999) and 17 millennial time scales (e.g. Mason et al., 2003).

18 Because of their nature, loess records are more challenging to interpret than marine or ice dust 19 stratigraphies in quantitative terms, but they hold great potential under opportune 20 circumstances. In the case of loess/paleosol sequences, the assumption is often made that EC 21 = 1, because the other soil component i.e. the organic matter content is usually very low i.e. 22 <1% (e.g. Miao et al., 2007). Nonetheless in carbon rich soils where the organic matter can be \sim 10%, this should be taken into account (Muhs et al., 2013). Therefore, the implication is that 23 the dust MAR is entirely determined by SBMAR = LSR * DBD. Depending on the study 24 25 DBD is either measured or assumed based on literature surveys, which adds significant uncertainty to calculations. The LSR is determined based on the age-depth relation. For this 26 27 compilation, focused on the Holocene, we only consider profiles were absolute ages (or more 28 correctly, numerical ages) have been measured, rather than relying on stratigraphic 29 correlations.

30 Depending on the availability of suitable material at loess sites, radiocarbon dating is carried 31 out on different organic components such as plant material (e.g. charcoal, plant and wood 32 fragments) and/or, or *Succineidae* (land snails). Humic acid is also utilized, however, this

1 medium provides less reliable dates. Scarcity of organic samples could be a limitation for

2 chronologies relying on radiocarbon dating. An alternative category of methods for numerical

3 dating of loess deposits is the luminescence-dating group of techniques (Roberts, 2008). In

4 particular OSL dating of quartz grains with the Single Aliquot Regenerative (SAR) dose

5 protocol (Wintle and Murray, 2006) is considered to be quite robust (Roberts, 2008).

6 Bioturbation by faunal burrowing is an active process complicating the interpretations of soil 7 profiles, as indicated by stratigraphic age inversions. In addition human activities such as 8 agriculture may cause significant perturbations to the upper sections of soil profiles (Roberts 9 et al., 2001). Additional problems in the interpretation of soil profiles may arise in cases 10 where the origin of the loess is not primarily eolian, but rather the product or reworking of 11 local deposits (Kemp, 2001). We therefore, did not consider sections from areas where such 12 occurrence was identified.

13 Even when reworked origin can be excluded, it should not be taken for granted that the 14 DMAR = DF relation necessarily holds in the case of loess deposits. Conceptually, we can 15 imagine the process of dust emission and deposition in a regional setting as follows: dust 16 emanates from a source and starts to be deposited downwind at rates decreasing with distance 17 from the source (Fig. 3). A clear example of this is evident in the maps showing the spatial 18 variability of the thickness of last glacial Peoria loess deposits in North America (Bettis et al., 19 2003), or the loess deposition in the Chinese Loess Plateau (CLP) (Liu, 1985; Lu and Sun, 20 2000). Understanding the spatial scale of this process is essential.

21 Grain size data from sampling transects at various locations suggest that a sharp decrease in 22 DMAR immediately downwind of source areas is associated with a decrease in the size 23 distribution within 20-50 km, before a slower decline in DMAR and size takes place 24 (Chewings et al., 2014; Mason et al., 2003; Muhs et al., 2004; Winton et al., 2014), and then 25 slowly keeps on the same trajectory on broader spatial scales (Ding et al., 2005; Lawrence and Neff, 2009; Porter, 2001; Prins et al., 2007; Sun et al., 2003). It is evident then that bulk 26 27 (i.e. over the entire size range) DMARs from profiles located within a very short distance (i.e. 28 20-50 km) from the sources are not suited to provide a representative estimate of DF over a 29 broad spatial domain, unless the spatial scale of interest is very fine (Cook et al., 2013). This 30 has substantial implications for climate models and reconstructions of the mass balance of 31 global dust cycle in general, because a misinterpretation of the significance of bulk DMARs

32 can drive large overestimation of DF (Albani et al., 2014).

1 On the other hand it happens that sites located in close proximity to the sources have the 2 highest accumulation rates, allowing for better chances of obtaining high resolution profiles 3 that are of great utility in paleoclimate reconstructions. Thus, often some of the better-4 resolved sites, especially those having an adequate time resolution to show variability during 5 the Holocene, tend to be close to the sources.

6 After the steep decline in bulk DMAR close to the source areas, we can imagine the DF 7 blanketing over the surface of the Earth, slowly decreasing as the distance from the source 8 increases, but approximately homogeneous over a broad area at a coarse enough spatial 9 resolution (Fig. 3). In reality the DMAR is highly dependent on the local landforms, both for 10 accumulation and preservation of the deposited dust (Stevens and Lu, 2009). Thus loess 11 deposited on escarpments facing the wind direction may be favourable for an enhanced dust 12 deposition (Bowen and Lindley, 1977; Mason et al., 2003). More often erosion is a major 13 player, so that DMAR < DF. Upland sites are generally considered more suitable 14 geomorphological settings to recover well-preserved profiles of DF (Derbyshire, 2003; 15 Kohfeld and Harrison, 2003; Mason et al., 2003; Muhs et al., 2003a). Field examination of the 16 broad area where a profile was studied may provide evidence of erosion (Lu et al., 2006), i.e. 17 if the horizon's stratigraphy is not widely reproduced regionally, but in some cases evidence 18 for erosion is only available via detailed independent age models (Buylaert et al., 2008; 19 Stevens et al., 2008). In addition, supporting data from other proxies in the profile, i.e. bio- or 20 chemo-stratigraphy, can provide grounds to establish the degree of coherence of specific 21 sections (Marković et al., 2011).

22 2.4 Other paleodust archives: Lake sediments and Peat bogs

Beside loess/paleosol sequences other land archives carry the potential to preserve dust stratigraphies: lakes and ombrotrophic peat bogs. Both can be located at an opportune medium range distance between the source areas and the more remote oceanic and polar sites. In addition, the preservation of large amounts of organic matter involve the possibility of high-resolution radiocarbon dating, which is of great value especially for a period such as the Holocene (Muhs et al., 2003b; Marx et al., 2009; Le Roux et al., 2012).

While diverse in nature, lakes and peat bogs also share some common issues that generally need to be addressed in order to provide reliable paleodust profiles: the possibility of

1 quantitatively isolating remote from local dust deposition, and the basin-scale 2 representativeness of eolian DMARs compared to DF.

3 In some circumstances (when fluvial inputs and rain outwash can be excluded) lake deposits 4 can preserve reliable dust stratigraphies, with little or no unconformities and relatively 5 abundant organic matter for radiocarbon dating (e.g. Muhs et al., 2003b). Maar lakes 6 developed in craters formed by explosive excavations associated with phreatomagmatic 7 eruptions, are often an ideal setting, when the mafic composition of the basin is substantially 8 different than the mineralogical and geochemical characteristics of the remotely originated 9 dust. However, a major problem with lakes is the possibility of sediment focusing in the 10 deeper parts of the basin, which may substantially affect SBMAR. With one exception, we 11 were not able to retrieve adequate DMARs from lakes for this compilation, mostly because of 12 problems with the age model, or a reliable estimation of EC (Supplementary material).

13 In recent years substantial progress was made in recovering dust profiles from ombrotrophic 14 peats. Estimation of SBMAR depends on the radiocarbon dating of the organic matter. The 15 EC is determined by the elemental composition of the residual ash after combustion of the 16 organic matter. The identification of an adequate proxy for dust can be challenging (Kylander 17 et al., 2013), so that several approaches including multi-proxy based approaches have been 18 suggested (Marx et al., 2009). Even more challenging is a quantitative isolation of the local 19 versus remote dust input, also because of the lack of size distribution data in most cases, 20 although a few studies have provided good approaches (Marx et al., 2009; Le Roux et al., 21 2012). At this stage, substantial uncertainties still exist in general in peat bog dust records for 22 one or more of the variables necessary to determine a reliable quantitative estimate of dust 23 MARs relevant for medium/long range transport. Nonetheless we expect that in the near 24 future this goal will be achieved, because of the fast progress of the research in this field (e.g. 25 Ferrat et al., 2011; Kylander et al., 2013; Marx et al., 2009; McGowan et al., 2010; Le Roux et al., 2012; Sapkota et al., 2007; De Vleeschouwer et al., 2012). 26

27

28 3 Methodology

The goal of this compilation is to provide a quality-controlled dataset with specific reference to the possibility of deriving reliable quantitative time series of eolian DMAR relevant to broad spatial scales. According to this principle and considering the specific characteristics of the different paleodust archives, we performed an extensive literature review to identify

records suitable for the study of dust variability within the Holocene, encompassing the MH 1 2

period ~6 ka BP.

3 There is a spectrum of possible approaches for the compilation of this kind of database, 4 comprised between two extremes: a minimal collection of DMARs (e.g. similar to 5 DIRTMAP, Kohfeld and Harrison, 2001), and an extensive compilation including a wide variety of metadata (e.g. DIRTMAP3, Maher et al., 2013). For this work, we lean towards the 6 7 first approach, although we include uncertainties and some additional information, but stick to 8 the age models from the original studies (Appendix A).

9 The concise operational product of the database is a set of dust MAR time series, with 10 quantitative estimates of the uncertainties associated to both the age and DMAR. Dust MAR 11 uncertainty quantified here is only associated with the calculations, hence it includes the 12 analytical errors and the uncertainty associated with assumptions or approximations in the 13 magnitude of specific variables. We express all quantitative uncertainties as 1^o deviation, 14 assuming a Gaussian distribution of the error. It will be expressed either in absolute terms or 15 as a relative error, as specified in each case.

16 This approach does not convey the overall uncertainty related for instance to a specific 17 technique or to a specific physical setting, which is difficult to express quantitatively. For this 18 reason we complement the dataset with a categorical attribution of the overall confidence on 19 the reliability of the records for the purposes of this work.

20 Note that a large part of the actual uncertainties associated with each record are related to 21 what we include in the attribution of the confidence level, and that the estimates provided for 22 the quantifiable uncertainty constitute a first order approximation.

23 In the following paragraphs we report the criteria followed for site selection and attribution of 24 a confidence level (Sect. 3.1), and we provide a general description of the approach used to 25 report or calculate the age profiles of eolian DMAR, with relative uncertainties (Sect. 3.2 and 3.3), and the information on the size distributions where available (Sect. 3.4). More specific 26 information for each record is reported in the Supplementary Material. In Sect. 3.5 we 27 28 describe the approach to estimate the mass balance of the global dust cycle throughout the

29 Holocene with the CESM.

1 3.1 Site selection and attribution of confidence level

- 2 In an initial phase of scrutiny of the existing literature we identified paleodust records of
- 3 interest to our project, based on the requirements that they:
- 4 a) have potential for calculating DMAR (i.e., the dust fraction must be identified and 5 quantified in some way; no records with only size information)
- 6 b) have sufficient material within the Holocene to quantify DMAR (i.e., at least three data
- 7 points occur between 0 and 11.7 ka BP, with at least 1 data point between 4.5 and 7.5 ka BP;
- 8 three data points means three ages for loess/paleosol sequences where EC = 1, and three
- 9 values of dust MAR for all other cases)
- 10 c) have absolute (i.e., numerical) ages (only for terrestrial sediments)
- 11 d) include size information (only for the loess/paleosol records)
- 12 We identified <u>124</u> sites meeting these criteria. We then labelled each of those sites with a
- 14 quantitative profile of eolian DMAR with respect to the age, and that it is relevant to broad

categorical attribution of the overall confidence we have that each record provides a

15 spatial scales, based on general consensus.

13

- 16 The attribution of the confidence level is based on whether or not there are substantial or
- 17 critical uncertainties with respect to three aspects: (1) SBMAR (and confidence that DMAR =
- DF); (2) EC; (3) quantitative distinction between remote and local EC (See SupplementaryTable 1).
- The first criterion (1) is related to the chronology itself, and/or linking the chronology to SBMAR. We consider some types of dates more reliable than others in this context, depending on the kind of natural archive. Among the less reliable, some we consider acceptable per se ("substantial uncertainty"), while others we associate with a "critical uncertainty".
- For marine sediments, we consider both absolute ages, and stratigraphic correlation with oxygen stacks, with the consideration that they are both acceptable in the case of records based on thorium profiling, but only absolute ages are acceptable when isolation of the terrigenous fraction is the method of determination of EC.
- For ice cores, we regard age models based on a combination of absolute counting, stratigraphic correlations, and ice thinning modelling (e.g. Veres et al., 2013) with high

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1 confidence. These models apply to most of the polar ice cores. On the other hand, records

2 from smaller ice caps and glaciers suffer from the lack of reliable age models, hence ice

3 accumulation profiles, which cannot be resolved on Holocene time scales at present (L.

4 Thompson, P. Gabrielli, C. Zdanowicz, personal comm.).

For terrestrial sediments, we only considered numerical ages (OSL, ¹⁴C), in the initial scrutiny 5 phase. This is important as in the case of loess/paleosol sequences, disturbances such as 6 7 erosion and reworking (and agricultural practices, when they are not limited to depths 8 attributed to the last ~ 2.5 ka) can disrupt the ideal correspondence between dust MAR and DF 9 (Sect. 2.3). We consider evidence of such an occurrence as a critical uncertainty. In addition, 10 we have attempted to identify sites whose stratigraphies are consistent regionally and 11 therefore demonstrate that they are more likely to represent large-scale patterns. Sites with 12 stratigraphies that diverge substantially from standard regional profiles suggest that these 13 records are not likely to represent large scale patterns in dust deposition, and this represents a 14 critical uncertainty. When no critical uncertainties are identified, we still consider that 15 SBMAR estimates from loess/paleosol sequences contain substantial uncertainty, according to 16 this criterion (1).

17 The second criterion (2) relates to the ability of a quantitative determination of the EC.

18 For marine cores, we rely on the original and subsequent authors' evaluation of 19 contamination, e.g., the possibility of non-eolian inputs such as from sediment focusing, 20 volcanic, fluvial, hemipelagic, and ice-rafted materials. Marine records that are definitely or 21 very likely to be affected by unaccounted for non-eolian inputs are rated as having critical 22 uncertainty. These include sites in regions that have been identified as being affected by non-23 eolian inputs such as the volcanic materials and ice-rafted detritus in the North Pacific (Serno 24 et al., 2014), volcanic inputs in the Eastern equatorial Pacific (Olivarez et al., 1991), possible 25 non-eolian detritus in the Western Pacific / Ontong-Java plataeu (Kawahata, 199), or sediment 26 focusing and Ice Rafted Debris (IRD) in the Southern Ocean (Kohfeld and Harrison, 2001). 27 When the possible presence of non-eolian components is more speculative, we attribute a 28 substantial level of uncertainty. In addition, estimates of EC made using quartz concentrations 29 or elemental (e.g. Al) proxies were rated as having substantial uncertainty. Records based on 232 Th, experimental isolation of eolian component, or a differencing method (EC = 1 - CaCO3 30 31 - opal - Corganic) to determine EC were preferred.

For ice cores, primary non-eolian inputs to the insoluble particle material are volcanic in 1 2 origin, and can usually be singled-out and selectively removed from the records (Narcisi et 3 al., 2010). In some cases though, they may be a widespread presence in a record (Gabrielli et 4 al., 2014), which we consider cause for attribution of substantial uncertainty. We consider 5 particle counters the more robust methods for the determination of EC. Un-calibrated (for the size) laser counters give unreliable results, as both the size distributions and the EC may be 6 7 significantly affected, which we consider a critical uncertainty. Among the 124 records-8 initially selected, a few ice core records rely on calcium as proxy for dust. Subtleties include 9 that total calcium is a worse proxy than non-sea salt calcium, and that calcium in general is a 10 better proxy in Greenland than in Antarctica, because the proportions of crustal versus nss-Ca in the two cases, with a sea salt deposition one order of magnitude higher than dust in 11 12 Antarctica, but much lower in Greenland (Ruth et al., 2002, 2008). We simply assume a 13 substantial uncertainty for all records based on calcium.

For terrestrial records, we attribute <u>substantial</u> uncertainty to the presence of non-eolian inputs, as identified by authors. We attribute substantial uncertainty when an elemental proxy

16 was used for the determination of EC, rather than relying on the sedimentation rate of the

eolian sediment, or the residual fraction after elimination of non-eolian inputs. A criticaluncertainty is attributed to the use of quartz as a quantitative proxy for EC.

- 19 The third criterion (3) focuses on the quantitative and size-resolved separation of local versus20 remote dust.
- 21 This criterion in fact does not apply to loess/paleosol sequences, where instead we had 22 applied constraints on the necessity of size information. For the other types of natural
- 23 archives, all the other records that we found to be most likely affected by unaccounted for
- 24 local dust inputs, are rated as having critical uncertainty. When the possible presence of local
- 25 dust inputs is likely, but more speculative, we attribute a substantial level of uncertainty.

Records that meet all criteria are labelled with "high confidence", whereas failing to meet one criterion results in a record receiving the attribution of "medium confidence" level. A record is given a low level of confidence when either (a) two or more aspects are considered affected

is given a low level of confidence when either (a) two or more aspects are considered affected
by substantial uncertainty, or (b) even one aspect is considered a critical uncertainty. We

30 included in the compilation only records (45) out of 124) with high and medium confidence

31 levels (Table 1; Supplementary material).

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1 3.2 Ages and chronologies

All the ages reported in this compilation are expressed in thousands of years before 1950 AD (ka BP). We do not re-derive the age models for the records in this compilation, but use the original chronologies reported in the relevant publications. This is the case for all records included in this compilation. The only exceptions are the case of the Antarctic ice cores, which have been reported to the AICC2012 chronology (Veres et al., 2013), and a specific approach for loess/paleosol sequences described below.

8 In the previous Section (3.1) we explained how loess/paleosol sequences with a medium 9 confidence level satisfy the condition of being representative of large scale patterns. This is 10 based on the possibility of grouping them within sub-regional settings where sequences 11 exhibit a common stratigraphy. These groups should also account for spatial variability in the timing of the onset of climatic conditions that are linked to specific loess/paleosol sub-units, 12 13 e.g. on the CLP. When possible (i.e., for the records in the Western CLP: Duowa and 14 Jiuzhoutai), we constructed SBMAR records for those sites, based on selecting (or 15 interpolating, in the case of Duowa: see Supplementary material) only the dates at the 16 interface between two consecutive sub-units, in fact reflecting the alternation of soil and loess 17 sub-units (S0.S1 - S0.L1 - S0.S2 - S0.L2 - S0.S3). We consider this as a slightly conservative 18 approach, which has the advantage of (a) limiting potential abrupt fluctuations in DMARs, 19 which may just be reflecting dating errors (e.g., related to bioturbation), and (b) pairing to 20 some extent the records, consistently with the criteria mentioned earlier. Note that a similar 21 approach was used for the two loess/paleosol sequences from Nebraska included in this 22 compilation (Wauneta, Logan Roadcut). For Jingyuan and the central CLP (Beiguoyuan, 23 Xifeng, Luochuan, Weinan), no such distinction of sub-units within the Holocene paleosol (S0) is visible, thus the time series are based on all the available dates. The same holds for the 24 25 one single site in Alaska (Chitina).

In the previous Section we discussed how either a linear or a more sophisticated age model is used to determine a profile's chronology. Each numeric age or tie-point is characterized by some uncertainty. The nature and magnitude of the error depend on the specific technique, and include the analytical error, and the calibration or wiggle-matching error when applicable. We try to estimate quantitatively this type of uncertainty. Unquantifiable uncertainties include the effects of bioturbation, sample contamination, etc.

Age uncertainties that can be estimated arise from 3 different processes: (1) experimental error in a measurement (e.g. ¹⁴C, OSL, etc.); (2) calibration errors (e.g. ¹⁴C calibration software; OSL measurement in water content); (3) other age model uncertainties. For instance radiocarbon dating requires corrections to account for the carbon reservoir effect (Brauer et al., 2014). Calibration software has been developed to perform this task (e.g. Bronk Ramsey, 1995; Reimer et al., 2009). All radiocarbon ages reported in this paper are calibrated, according to the original references.

8 In the case of age models more complicated than the simple liner relation used to derive a 9 LSR, errors associated with ages are usually reported in the publications. An example of this 10 are the new ice core chronologies, such as AICC2012, which report the associated age 11 uncertainties (Veres et al., 2013).

For a linear sedimentation model, the age of a given depth horizon is calculated by linear interpolation between two dated horizons. In this case the age error of the samples is bound to the uncertainties associated with the bracketing ages. The age-model error of the sample can then be derived through the error propagation formula:

16
$$\varepsilon_{sample} = \sqrt{\varepsilon_a^2 + \varepsilon_b^2}$$
 (1)

17 where ε_a and ε_b are the age errors of the two adjacent dated points between which the linearly 18 interpolated sample age was calculated.

19 The other usual possibility is that the age model of a site was determined without the help of 20 any absolute age marker, but just using stratigraphic correlation. A typical example of such an 21 age model is one based on stratigraphic correlation of a marine sediment core site's $\delta^{18}O$ 22 profile with the SPECMAP stack (Imbrie et al., 1984). In this case and in all other 23 circumstances where the age error is not reported, we arbitrarily assume an uncertainty of 24 6.8% (1 σ , corresponding to an overall 10%).

25 3.3 Eolian Dust MARs

Dust MARs constitute the key element of this compilation. We previously discussed (Fig. 2) the non-parallel depth resolution of the age samples and the EC samples. Unless stated otherwise, we always use a chronology targeted on the final DMAR resolution, which is determined ultimately by the EC resolution (see also Fig. 1). The typical exceptions are

loess/paleosol sequences, where SR alone (hence the resolution of the age samples)
 determines the dust MAR.

We report both the SBMAR (or SR and DBD) and EC for each point in the records, with relative uncertainties. The uncertainties are taken from the original sources when available, and assigned otherwise. The dust MAR uncertainty is determined from the relative uncertainties in the factors SBMAR and EC, combined through the error propagation formula:

7
$$\varepsilon_{MAR} = \sqrt{\left(\frac{\varepsilon_{SBMAR}}{\mu_{SBMAR}}\right)^2 + \left(\frac{\varepsilon_{EC}}{\mu_{EC}}\right)^2}$$

8 with $\varepsilon_{SBMAR/EC}$ and $\mu_{SBMAR/EC}$ representing the absolute errors and the <u>absolute values</u>, 9 respectively.

In this compilation, there are two cases when SBMAR is provided directly instead of being 10 the combination of SR * DBD: ice cores and marine sediment records derived using the 11 12 thorium profiling method. In the case of ice cores SBMAR corresponds to the ice 13 accumulation rate, expressed in m (water equivalent) per year, which incorporates information 14 about ice density and thinning with depth (Alley, 2000; Veres et al., 2013). When not 15 reported, we assume that the relative uncertainty is the same as that of the age uncertainty. 16 This is a reasonable approximation for the Holocene records from the ice cores presented 17 here, but significantly larger uncertainties related to ice thinning models should be considered 18 for deeper sections of ice cores and for glacial stages (Kindler et al., 2014). For marine cores, 19 we consider the relative uncertainty in the thorium excess (xs-Th) parameter. When not 20 reported we assumed a relative uncertainty of 5%, assigned based on an expert informed 21 guess.

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In all other cases, for SR we consider that the relative uncertainty is the same as the age uncertainty, which again is combined through the error propagation formula to the other uncertainties. DBD is sometimes measured, often just assumed based on the literature from the broad region. When no information was reported in the original works, we assumed a dry bulk density of 1.48 g cm⁻³ for the CLP (Kohfeld and Harrison, 2003), and 1.45 g cm⁻³ for North America (Bettis et al., 2003). When not measured, we assumed 15% relative uncertainty for DBD (Kohfeld and Harrison, 2003).

With the exception of loess, for which we assume EC = 1 unless otherwise stated, EC is either expressed in terms of fraction or concentration of dust or a proxy in the bulk sediment. For the

23

(2)

- 1 Antarctic ice cores considered in this compilation the EC is determined after the volume dust
- 2 concentrations determined by a Coulter Counter; the mass concentration is calculated by
- 3 multiplying that per the assumed dust density of 2.5 g cm⁻³ (Delmonte et al., 2004). The
- 4 uncertainty in this case is taken from the standard deviation of the ~3 replicate measurements.
- 5 When a dust proxy is used instead to determine the EC, its concentration is divided by the
- 6 element's typical abundance in dust (or crustal abundance). In this case the analytical
- 7 uncertainty (if not reported, we assume 5%) is combined with the uncertainty of the dust
- 8 proxy i.e. the variability of its amount in dust. We keep the proxy-dust relation from the 9 original studies when available.
- Several records in this compilation use ²³²Th as a dust proxy, for which we assume 10.7 ppm in dust (McGee et al., 2007) if not specified otherwise in the original papers. We always assumed 9.3% uncertainty for ²³²Th as a dust proxy (McGee et al., 2007), or a combined uncertainty of 15% when the analytical uncertainty was not available. In one case (GISP2) we used calcium as a dust proxy (Mayewski et al., 1997), assuming a variable calcium-dust relation in Greenland with climate conditions, resulting in 26% calcium in dust (Ruth et al., 2002; Steffensen, 1997), with an arbitrarily assigned uncertainty of 20%.
- When isolation of the detrital component from the sediment matrix is done by removal of carbonates, opal, and organic matter, then the EC can be estimated from the bulk terrigenous component. We assume 5% uncertainty in this procedure.
- We stress once again that the quantitative uncertainties estimated here do not fully represent the overall uncertainty of a record, which should be pondered in combination with the confidence level (Table 1).

23 3.4 Dust grain size distributions

Here we focus on the importance of the grain size information and its intimate link to the DMAR. When possible, we retrieved the size distributions associated to the records in this compilation. Depending on the technique used, the size data was collected in the form of size distributions (e.g. particle counters and laser particle analysers) or size classes (sieve and pipette method), e.g. the percentages of sand, silt, and clay (Muhs et al., 2013; Lu et al., 1999).

- 30 Despite the differences and uncertainties associated with specific methods (Mahowald et al.,
- 31 2014; Reid, 2003), we include the available information according to the original sources. In

1 the case of size classes, we report the information as provided in the original papers. In 2 addition, we take an innovative approach to organizing the size distribution data. First of all, 3 we carry the original size distributions to a new, common binning, in order to enhance the 4 accessibility of the data and to facilitate the inter-comparison among records. Second, we 5 associate the size distributions to the DMAR time series sample-to-sample where possible, so 6 that DMAR time series for different size ranges can be easily determined.

The re-binning procedure to adapt the original size distributions from observations is organized in a series of steps: (1) definition of a new binning model; (2) building the cumulative distribution from the normalized observations; (3) fitting a spline curve to the observation cumulative distribution; (4) integration of the fitted spline curve into the new bins; (5) evaluation and summary of the fit of the new binned data to the original observations. The fitting spline in (3) is bounded to have values between 0 and 1, and to be monotonically non-decreasing.

14 One challenge in finding a new binning model is to avoid significant distortion to the original 15 size distribution, given that observations have both a different resolution and a different size 16 range. A compromise is necessary to preserve both the actual dust flux (i.e. a size range wide 17 enough to embrace most observations) and the shape of the distributions. Preservation of the 18 size distribution properties, i.e. the mass partitioning across the size spectrum, requires an 19 adequate number of bins and adequate spacing. We adopted a new bin model with n = 7620 bins, spanning the interval of particle diameters between 0.28 and 208.34 µm. The bin spacing is defined by a monotonically increasing function: y = 0.089 * x + 0.002, where x is the nth 21 bin centre, y is the $(n+1)^{th}$ bin centre, and $x0 = 0.35 \ \mu m$ (first bin centre). Bin edges are 22 23 calculated by linear interpolation, halfway between two consecutive bin centres. This binning 24 model is very similar to the instrumental size binning of e.g. Mulitza et al. (2010) or McGee 25 et al. (2013) in the same size range. For all samples subject to re-binning, visual inspection of the original and new distributions was performed, as well production of objective metrics 26 27 (Supplementary material).

All references to the size in this work refer to the particle's diameter. We always refer to
volume/mass size distributions, both in the main text and the Supplement.
1 3.5 Modelling the global dust cycle

Paleodust records not only represent excellent climate proxies, but they also offer the 2 3 possibility to quantitatively constrain the mass balance (or magnitude) of the global dust 4 cycle. Here we use a dust model to extrapolate the available data to allow global coverage for 5 the deposition, as well as estimates of sources, concentrations and aerosol optical depth using 6 the Community Earth System Model (Albani et al., 2014; Mahowald et al., 2011, 2006). To 7 represent the impact of climate variability during the Holocene onto the dust cycle, we chose 8 two reference periods for our simulations with the CESM: the MH (6 ka BP) and the pre-9 industrial (1850 AD), which we assume representative for the Early and Mid-Holocene (5-11 10 ka BP) and the Late Holocene (1-5 ka BP) respectively, based on the first-order differences in 11 orbital forcing and climate in the two periods (e.g. Wanner et al., 2008). The initial conditions 12 for the MH simulations are taken from a fully-coupled climate equilibrium simulation for 6 ka BP (http://www.cesm.ucar.edu/experiments/cesm1.0/#paleo), which follows the PMIP3 13 14 prescriptions for greenhouse gases concentrations and orbital forcing, with pre-industrial 15 prescribed vegetation (Otto-Bliesner et al., 2009), and was part of the PMIP3/CMIP5 model 16 experiments for the IPCC AR5 (Masson-Delmotte et al., 2013; Flato et al., 2013). For the pre-17 industrial simulation we take the initial conditions from an equilibrium reference simulation 18 described in Brady et al. (2013). 19 The dust model integrated in the CESM used for this study uses the Community Atmosphere 20 Model version 4 with a Bulk Aerosol Model (CAM4-BAM), and is described in detail in

Albani et al. (2014). The dust model simulates dust emission, transport, dry and wet deposition, and direct interactions with radiation in the long and shortwave spectrum. The dust mass is partitioned in four size classes spanning the 0.1-10 μm diameter range. Modelled dust emissions are primarily a function of surface wind speed, vegetation (and snow) cover, and soil erodibility, which is a spatially-varying parameter summarizing the differences in susceptibility to erosion related to e.g. soil textures and geomorphology (Zender et al., 2003).

Although the physical model does not include changes in vegetation, following the PMIP protocols (Otto-Bliesner et al., 2009), we accounted for different vegetation cover in the MH by removing the online dependence of dust mobilization on preindustrial vegetation. For the 6 ka BP equilibrium climate instead we simulated new vegetation cover with BIOME4 (Kaplan et al., 2003), following the methodology of Mahowald et al. (2006). The effects of vegetation were incorporated in the soil erodibility map by applying a scale factor at each grid cell,

1 proportional to the fraction of the grid cell available for dust emission in arid areas (same as

2 for the LGM in Albani et al., 2014). We also accounted for glaciogenic sources in Alaska,

3 which are not explicitly simulated by the model, by prescribing them according to Albani et

4 al. (2014) and Mahowald et al. (2006).

5 In addition, we relaxed the dampening effect of vegetation cover on dust mobilization in the 6 model in one specific region, i.e. the Nebraska Sand Dunes, to account for a known dust 7 source relevant for the Holocene (Miao et al., 2007). In that region, too much vegetation 8 cover from the prescribed input datasets would otherwise inhibit dust mobilization for both 9 for the pre-industrial and MH simulations.

We provided observational constraints on the model dust deposition flux by considering the dust MAR from the data compilation, limited to the model's size range i.e. $\leq 10 \mu m$: we considered only the relevant fine fraction from the new binning. For each record we calculated MAR time series during 2 ka-long time intervals centred on 2, 4, 6, 8, and 10 ka BP, by averaging the original data across each of the macro-regions (Fig. 4). Linear interpolation was then used to fill-in the gaps.

16 The model's fit to the observations was improved through a spatial optimization of the soil 17 erodibility, by applying a set of scale factors specific to macro-areas, which is reflected on 18 dust mobilization from those macro-areas (Albani et al., 2012b, 2014; Mahowald et al., 2011, 19 2010, 2006). We applied this procedure to pre-industrial and MH simulations constrained by 20 the data in the 4 ka BP and 6 ka BP time slices, respectively. In order to account for dust 21 variability in the other time periods (2, 8, and 10 ka BP) we linked them to the respective 22 reference case for the Late (4 ka BP) and Mid/Late Holocene (6 ka BP), by prescribing an 23 additional set of scale factors for dust emissions in the same model macro-areas. Those scale 24 factors are expressed as anomalies to the reference period, and are determined based on the 25 observations: each time series in the compilation at the 2 ka pace was reduced to an anomaly 26 with respect to its value at 6 ka BP (and 4 ka BP in parallel), then a regional average anomaly 27 was calculated within specific regions determined based on the geographical distributions of 28 the observations (Fig. 4). We assume that emissions in each of the model macro-areas are 29 related to observations from specific geographic regions, which act as sinks for dust originated from each dust source macro-area (Mahowald et al., 2010). The anomaly in dust 30 31 emissions was then calculated as the average of the anomalies from the group of forcing

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1 regions (Table 2). We acknowledge that this simple procedure implies possible discontinuity

- 2 at the 4 to 6 ka BP ka BP transition.
- 3

4 4 Holocene dust variability

5 4.1 Global overview

- 6 A total of $\frac{45}{124}$ high- and medium-confidence paleodust records (out of $\frac{124}{124}$) from ice, terrestrial
- 7 and marine archives distributed worldwide comprise the data compilation (Fig. 5). It is
- 8 noteworthy that while in a few regions there is a relative abundance of observations (North

9 Atlantic, Equatorial Pacific) there are few data from other parts of the world (North Pacific,

10 Southern Hemisphere), after the application of filtering criteria.

11 The amplitude of bulk dust variability recorded from natural archives during the last 22 ka

12 relative to their Holocene average allows a comparison with the DIRTMAP3 (Maher et al.,

13 2010) data with regard to the glacial / interglacial variability within several regions around the

14 globe (Fig. 5).

15 Different regions show different patterns of variability during the Holocene (e.g. the apparent

16 little variability in the Equatorial Pacific versus the Mid-Holocene minimum in the North

17 African margin), and even within certain regions there may be significantly diverse trends,

18 which will be discussed in more detail in the following Sections.

19 4.2 North Africa and North Atlantic

The most striking display of variability during the Holocene is shown by the cores in North-20 21 western African Margin (13 records), with an amplitude comparable to glacial/interglacial variability (Fig. 5) (Adkins et al., 2006; McGee et al., 2013). As first suggested by deMenocal 22 23 et al. (2000), this would be a clear mark of the significant changes in the climatic conditions 24 in North Africa between the wetter Early to Mid-Holocene compared to the drier late glacial 25 and Late Holocene. During the so called "African Humid Period" in the Early to Mid-Holocene, greening of the Sahara occurred i.e. changes in vegetation in response to increased 26 27 humidity and precipitation, as seen in pollen records and lake level changes (e.g. Hoelzmann 28 et al., 1998; Jolly et al., 1998; Street-Perrott and Perrott, 1993). The cause of these changes 29 has been identified as an enhanced summer monsoon, driven by changes in orbital forcing,

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1 sea surface temperature and vegetation changes (e.g. Braconnot et al., 2007; Claussen et al.,

2 1999; Kutzbach and Liu, 1997).

3 Figure 6 shows the large range of values (spanning two orders of magnitude) encompassed by

4 the DMAR estimates from marine sediment cores to the west of the African coast. Records

5 from the Equatorial Atlantic (lower temporal resolution) tend to show decreasing trends from

6 the Early to Mid-Holocene, with little or no variability afterwards (Bradtmiller et al., 2007;

7 François et al., 1990), compared to the sites on the NW African margin (higher temporal

8 resolution) that show a minimum in DMAR in the ~5-9 ka BP period (McGee et al., 2013).

9 The absolute values of bulk DMARs (dotted lines) are higher for the sites close to the coast of 10 NW Africa (bluish colours) compared to the sites in the Equatorial Atlantic (reddish and 11 greenish tones). When considering only the fine fraction ($< 10 \mu$ m: solid lines), 3 (out of 5) 12 records from the NW African margin are comparable in magnitude to those in the equatorial 13 Atlantic, at least for the Early to Mid-Holocene, but tend to be larger in the Late Holocene. 14 On the other hand, 2 of the records display very low values of DMARs, lower than the 15 records from the equatorial Atlantic, and comparable to the Equatorial Pacific.

Core-top bulk dust MARs from NW African margin cores match very well with modern sediment trap data (Ratmeyer et al., 1999). On the other hand there is substantial uncertainty in the attribution of the fine fractions, with records in the Equatorial Atlantic loosely constrained by present-day sediment trap data from the Cape Verde area (Ratmeyer et al., 1999), and size data for the NW African margin based on actual measurements from sediment samples, but relying on end-member modelling for the separation between riverine and eolian inputs (McGee et al., 2013).

This compilation and comparison suggests that there is still a substantial knowledge gap in the area, and ample space to debate the causes of the differences in magnitude and trends between the records form the NW African margin and the Equatorial Atlantic. For instance, there could be differences, related to shifts in the position of the ITCZ with relation to the dust plume, or to differences in the interpretation of the data, in particular with reference to the grain size distributions and potential non-eolian components, with implications for the spatial representativeness of the records.

1 4.3 Arabian Sea

2 Marine sediments from the Arabian Sea are of great value, as they provide a rare opportunity 3 to gather information about past dust variability from the Middle East and Central Asia, from 4 which little is known despite this arid belt being one of the major dust sources worldwide 5 (Prospero et al., 2002). The most relevant climatic feature in the region is the seasonality 6 related to the onset of the SW Indian monsoon. The largest dust activity in the region is from 7 summer dust emissions from Mesopotamia and the Arabian Peninsula, which are thought to 8 constitute the major dust sources at present for the Arabian Sea, although contributions from 9 Somalia and Iran / Pakistan may be important (Prospero et al., 2002).

10 We report data from the cores RC-27-42 and 93KL, recovered from the central Arabian Sea

11 (Pourmand et al., 2007) and the Little Murray ridge in the Northeast (Pourmand et al., 2004),

12 respectively. There are no clear common trends between the two records, which indeed show

13 very different DMARs, one order of magnitude apart (Fig. 7). There is little information to

explain the difference in magnitude, which is perhaps related to different sources, although possible fluvial inputs to 93KL cannot be conclusively ruled out. There is clear evidence that

16 dust grains larger than 10 μm are present in the Arabian Sea sediments (Clemens and Prell,

17 1990; Clemens, 1998; Sirocko et al., 1991). The fine fraction ratio for the two records is a

18 rough approximation common to both records (Table 1).

19 4.4 North America

Evidence of dust deposition and accumulation during the Holocene in North America is widespread, mainly linked to loess deposits in the mid-continent (Bignell loess), particularly in Nebraska (Mason et al., 2003; Miao et al., 2007), Kansas (Feng et al., 1994), North Dakota (Mason et al., 2008), and Eastern Colorado (Muhs et al., 1999; Pigati et al., 2013). Most areas are characterized by relatively low thickness, so that low temporal resolution does not allow assessing Holocene variability, with the exception of a few sites in Nebraska (Miao et al., 2007).

Unlike the other areas where loess origin is related to local river systems, loess deposits in Nebraska have their immediate sources in the extensive dune fields to the Northwest. Changes in the climatic conditions affecting vegetation cover have the potential to loosen or stabilize the dunes, altering their potential as dust sources (Miao et al., 2007).

1 Well-studied sites at Wauneta have very high temporal resolution due to the high DMARs,

2 and allowed the identification of different phases of dust accumulation and pedogenesis

3 during the Holocene. The high accumulation rates are related to the location, on the edge of

4 tableland escarpments facing the immediate source areas of the dust. The accumulation rate

5 drops off drastically in the downwind direction from these sites, for example, the \sim 6 m of

6 Holocene loess in the Old Wauneta section thins to a little over a meter within a few hundred

7 meters downwind, where a rather uniform loess mantle covers the upland sites (Jacobs and

8 Mason, 2007; Mason et al., 2003). Another site to the NE (Logan Roadcut) shows lower bulk

9 DMAR but similar phasing, associated to the sequence of pedostratigraphic horizons (Miao et

10 al., 2007). When accounting for the size information i.e. when focusing on the fine fraction

11 DMARs, both the absolute values of DMAR drastically decrease, and become comparable in

12 magnitude (Fig. 8). This suggests that the fine fraction DMARs can be considered more

13 representative (rather than bulk DMAR) of accumulation rates over large areas.

14 4.5 Alaska

Dust activity in Alaska has been reported for both the present day (Crusius et al., 2011) and the past, in glacial and interglacial times (Muhs et al., 2003a). Dust in Alaska is of glaciogenic origin i.e. results from the formation of loose sediments characterized by fine particles, produced by the abrasion of the ice over the surface sediments or bedrock, and released on river/streams outwash plains during the melting season (Bullard, 2013).

Loess deposits of Holocene origin have been identified in central (Begét, 1990) and southern Alaska (Muhs et al., 2004, 2013; Pigati et al., 2013). The only site with high temporal resolution and numerical dating is the Chitina section in the Wrangnell-St. Elias National Park (Muhs et al., 2013; Pigati et al., 2013). The high bulk DMAR (Fig. 9) suggests that the dust sources (attributed to the Copper River basin) lay very close. This notion is supported by the coarseness of grain size data, comparable to analogous data from sites in the Matanuska Valley, which are located within 10 km from the putative source (Muhs et al., 2004).

Another record with Holocene temporal resolution is from a maar lake (Zagoskin Lake, on St.
Michel Island) in Western Alaska, which is thought to be representative of proximal but not
strictly local sources (Yukon River Valley), as also shown by the grain size (Muhs et al.,
2003b). When the fine fraction of DMAR is considered, the Chitina section and Zagoskin
Lake show comparable magnitude (Fig. 9), which we observe is rather large in a global

1 perspective. While this indicates that dust deposition into the Alaskan Gulf and other 2 surrounding seas is probably relatively large (Crusius et al., 2011), it is difficult to assess if 3 the spatial extent of Alaskan dust sources is such that the region is a quantitatively relevant 4 source for dust in the high latitudes (Bullard, 2013; Muhs et al., 2013). Geochemical tracer 5 studies in the North Pacific may provide some clue (Serno et al., 2014).

6 4.6 East Asia and North Pacific

7 The deserts in Western and Northern China are major global dust sources with relevance for the mid and high latitudes of the Northern Hemisphere (e.g. An et al., 1991; Lu and Sun, 8 9 2000; Bory et al., 2003; Prospero et al., 2002). The most stunning evidence of East Asian dust 10 history in the Quaternary and beyond in response to orbital forcing lies in the thick deposits of 11 the Chinese Loess Plateau (CLP), which covers extremely vast areas of the upper and middle 12 reaches of the Yellow River to the Southeast of the Badain Juran, Tengger and Ordos deserts (e.g. Ding et al., 2005; Kohfeld and Harrison, 2003; Kukla and An, 1989; Porter, 2001). In 13 14 relation to the vastness of the CLP, different climatic forcing mechanisms may have inter-15 played in a varying fashion in different regions, in response to changes related to the monsoon 16 system (Cosford et al., 2008, Dong et al., 2010), including in the extent or activity of the 17 source areas (e.g. Lu et al., 2013, 2010), in transport, i.e. wind strength and/or seasonality 18 (e.g. An et al., 1991; Ding et al., 2005), and climatic conditions controlling the balance of 19 pedogenesis and loess accumulation (e.g. Jiang et al., 2014).

20 Despite several studies conducted on the CLP, a few absolutely dated records exist that have 21 Holocene temporal resolution (Kohfeld and Harrison, 2003; Roberts et al., 2001), with some 22 additions in more recent years (Stevens et al., 2006, 2008, 2010; Lu et al., 2006, 2013). In 23 many areas agricultural practices carried out for at least the last ~2.5 ka complicate the 24 interpretations of the upper parts of several loess/paleosol sequences (e.g. Roberts et al., 25 2001). We selected two sites with loess/paleosol sequences from the Western CLP, from Duowa (Maher et al., 2003; Roberts et al., 2001) and Jiuzhoutai (Kohfeld and Harrison, 2003; 26 27 Sun et al., 2000). The two sites show the same sequence of pedostratigraphic succession of loess and paleosol sub-units (Kohfeld and Harrison, 2003; Roberts et al., 2001; Sun et al., 28 29 2000), and the bulk DMAR corresponding to the alternation of those sub-units show similar 30 trends (Fig. 10). When the fine component alone is considered, the DMARs from the two sites 31 are very similar. For those reasons, the two sites seem to be representative of large-scale 32 patterns in the Western CLP. We also report DMAR from another site in the western CLP

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(Jingyan: Sun et al., 2012), and from four sites located in the central CLP: Xifeng and 1 2 Beiguoyuan (Stevens and Lu, 2009), Luochuan (Lu et al., 2000, 2013), and Weinan (Kang et 3 al., 2013). Those sequences have similar soil unit stratigraphy for the Holocene (Sect. 3.2), 4 but the DMARs relative trends are not consistent (Fig. 10), possibly indicating that local 5 effects may have some more diffuse influence on DMARs in the central CLP sites. The central sites show a more uniform stratigraphy during the Holocene (prevalence of 6 7 pedogenesis) with respect to the sites in the Western CLP, possibly indicating a stronger 8 influence of the summer monsoon.

Dust plumes emanating from Asian deserts provide dust inputs to the Northern Pacific Ocean
(Rea, 1994), but because of low sedimentation rates and the lack of carbonate-rich sediments
the information from records with temporally resolved Holocene is very limited. We show
one record from core V21-146 (Hovan et al., 1991), which exhibits relatively little variability
during the Holocene.

14 4.7 Greenland

15 Ice core records from Greenland are among the best temporally resolved paleoclimate proxies. 16 They show the sharpest and largest amplitude oscillations observed in paleodust records 17 worldwide, following the trends exhibited by the other proxies such as e.g. δ^{18} O in the 18 alternation of stadial and interstadial phases during the last glacial period and the deglaciation 19 (Fuhrer et al., 1999; Mayewski et al., 1997; Ruth, 2003; Steffensen et al., 2008).

Among the ice cores drilled in Greenland, only one has a full Holocene dust record: GISP2 (Mayewski et al., 1997; Zdanowicz et al., 2000; Zielinski and Mershon, 1997), for which we considered the calcium record as a proxy for dust (Mayewski et al., 1997). Compared to the large variability of the glacial period, the Holocene dust MAR is rather flat, but a closer inspection shows an increasing trend from the Early to the Mid-Holocene, followed by a declining trend in the Late Holocene and a rise during the last millennium (Fig. 11).

It is not clear whether dust variability during the Holocene at GISP2 is related to (1) changes in the dust sources, which are thought to be in central and East Asia (e.g. Bory et al., 2003), (2) the atmospheric circulation, which indeed played a major role during the sharp glacial climate transitions (Mayewski et al., 2014; Meeker and Mayewski, 2002; Steffensen et al., 2008), or (3) changes in deposition mechanisms, which was suggested to be important on glacial/interglacial time scales but may be of minor relevance during the Holocene when Cornell University 3/5/15 10:01 AM **Deleted:** and

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1 accumulation rates are thought to be rather stable (Unnerstad and Hansson, 2001). New

2 studies spanning the Holocene perhaps using dust MARs from particle counters at other sites

3 may help understanding if this is a consistent feature of dust deposition in Greenland.

4 4.8 Equatorial Pacific

5 The Equatorial Pacific Ocean is one of the most remote regions in the world. It is 6 characterized by low dust deposition, correlated with global ice volume and dust in Antarctic 7 ice cores over glacial/interglacial cycles (Winckler et al., 2008). The spatial coverage in the 8 region is relatively good, in that there are North-South and East-West transects of cores with 9 temporally-resolved Holocene to Last Glacial dust records (Anderson et al., 2006; Bradtmiller 10 et al., 2006; McGee et al., 2007).

11 The sites consistently show larger DMARs during the Early Holocene compared to the MH 12 and Late Holocene (Fig. 12), with the two northernmost records from 110 W (green tones) showing the highest DMARs in then region. Due to the low sedimentation rates of equatorial 13 Pacific sediments (typically 1-2 cm ka⁻¹), it is uncertain whether these Holocene trends are 14 15 real or simply reflect bioturbative mixing of glacial sediments characterized by high dust 16 MARs with lowermost Holocene sediments. The records generally show decreasing DMAR 17 from North to South, and from East to West. Geochemical fingerprinting of dust in the Equatorial Pacific sediments indicates a complex situation, with a mixture of potential dust 18 19 sources including Asia, North and Central/South America, Sahara, and Australia (Xie and 20 Marcantonio, 2012; Ziegler et al., 2007).

21 4.9 Australia

Australia's drylands are among the largest dust sources in the Southern Hemisphere in the present day (Prospero et al., 2002), and dust deposits on land and in the surrounding seas archive evidence of the continent's dust history during glacial and interglacial cycles (De Deckker et al., 2012; Hesse and McTainsh, 2003; Lamy et al., 2014). The paucity of data for the Holocene in the Australian region was stated at the time of the DIRTMAP compilation (Kohfeld and Harrison, 2001), and since then more research was carried out (Fitzsimmons et al., 2013; Marx et al., 2009; McGowan et al., 2010).

We report (Fig. 13) two marine sediment records sampling the two main dust corridors emanating from Australia: the Tasman Sea (Fitzsimmons et al., 2013; Hesse, 1994), and the

East Indian Ocean (Fitzsimmons et al., 2013; Hesse and McTainsh, 2003). The NW core from the monsoon-influenced zone shows relatively high dust MAR during the Early Holocene and a declining trend toward the Mid- and Late Holocene (Fitzsimmons et al., 2013). On the other hand the core from the Tasman Sea shows a minimum dust MAR during the Early Holocene compared to the MH, in line with trends reported from a peat bog in New Zealand (Fitzsimmons et al., 2013; Marx et al., 2009).

7 4.10 South Atlantic Ocean

8 There is some information about lithogenic DMAR in the southern oceans in the literature, 9 but a quantitative estimation of eolian DMAR directly related to the atmospheric DF can be 10 problematic, because of low dust DF coupled with strong sediment redistribution by currents 11 and input of non-eolian material carried by floating icebergs i.e. ice-rafted debris (e.g. 12 Kohfeld et al., 2013). Nonetheless a few studies exploiting the thorium profiling method 13 attempted to correct SBMAR for sediment redistribution, providing new data (Anderson et 14 al., 2014; Lamy et al., 2014).

In particular the dust record from core PS2498-1 recovered from the Mid-Atlantic Ridge in the sub-Antarctic South Atlantic Ocean (Anderson et al., 2014) is characterized by high temporal resolution during the Holocene (Fig. 14). The dust, whose source is hypothesized to be from South America, shows a marked declining trend during the Holocene, with late

19 Holocene values a factor of ~ 2 lower than those found in the Early Holocene.

20 4.11 Antarctica

Ice core records from the East Antarctic plateau (Delmonte et al., 2004; Lambert et al., 2008) represent high quality dust records in terms of temporal resolution, reliability of the age model (Veres et al., 2013), isolation of the eolian component and measure of its size distribution (Delmonte et al., 2004, 2013), identification of remote sources (Albani et al., 2012b; Delmonte et al., 2010b) and broad scale spatial representativeness (Mahowald et al., 2011). Similar to Greenland, the Holocene dust MAR in the East Antarctic Plateau shows

27 little variability compared to the large glacial/interglacial and stadial/interstadial variations.

28 Both records considered in this study, EPICA Dome C (EDC) and Vostok-BH7 (Delmonte et

29 al., 2004; Lambert et al., 2008), show a slightly declining trend in dust MAR throughout the

30 Holocene, superimposed on large variability (Lambert et al., 2012) (Fig. 15). Some

- 1 deglaciated areas and nunataks at the edges of the ice sheets are prone to act as dust sources
- 2 (Bory et al., 2010; Bullard, 2013; Chewings et al., 2014; Delmonte et al., 2010b, 2013). In
- 3 such a remote environment, even small amounts of local dust can give a relevant contribution
- 4 to the dust budget of ice cores e.g. TALDICE (Albani et al., 2012a; Delmonte et al., 2010b).
- 5 Because dust from Antarctic sources does not travel in significant amounts to the interior of
- 6 the East Antarctic Plateau (Delmonte et al., 2013), it is unlikely that the declining Holocene
- 7 DMARs at Vostok and Dome C are related to the large variations seen in the TALDICE
- 8 record (Albani et al., 2012a).
- 9 Possible explanations may be related to the interplay of the contributions from different dust
- 10 source from South America and Australia (Albani et al., 2012b; Delmonte et al., 2010b), and
- 11 atmospheric circulation changes.

12 4.12 Mass balance of the global dust cycle throughout the Holocene

A detailed comparison of modelled and observed dust deposition ($<10 \mu$ m) for 6 ka BP (5-7 ka BP interval) is shown in Fig. 16 (see Supplementary material for the other time periods and the dominant sources). The modelled deposition is generally consistent with the observations of dust MAR spanning 6 orders of magnitude, within a factor of 10, similar to previous studies (Albani et al., 2014; Mahowald et al., 2006). Nonetheless, there are a few notable outliers.

19 While modelled deposition in the Equatorial Atlantic is very well reproduced, observations of 20 DMAR in the NW African margin appear to suggest overestimation by the model for some 21 sites in that region. There are several possible (perhaps concurrent) explanations worth 22 considering. First of all, the model may not be able to represent adequately the spatial 23 distribution of dust sources within North Africa, resulting in a different localization of the dust plume hence a different North-South gradient in the dust deposition. On the other hand, it 24 25 is possible that some inconsistencies exist among observations, due to different 26 methodological approaches, as discussed in Sect. 4.2. From a global perspective, there is an 27 interesting aspect emerging from Fig. 16, which may support this argument. The observational DMARs in some of the North African margin cores are comparable to or 28 29 smaller in magnitude than some of the cores in the Equatorial Pacific, which was unexpected, 30 and became evident once the size information was taken into account and coupled to the dust 31 MARs.

1 In addition to the possible methodological inconsistencies outlined above, two other potential 2 explanations for comparable fine (<10 μ m) DMARs on the NW African margin and the 3 equatorial Pacific could be: (a) a lack of wet deposition on the NW African margin, possibly 4 leading to low deposition of fine dust particles there, despite high atmospheric dust loads; (b) 5 possible substantial overestimation of dust deposition in the Holocene in the equatorial 6 Pacific, due to bioturbative mixing of glacial and Holocene sediments in this regions with 7 very low sedimentation rates (1-2 cm ka⁻¹).

8 We also note how South Atlantic DMARs are almost as large as the largest deposition rates 9 observed downwind of North Africa for the fine fraction, in a region where satellite images 10 show little dust loading today (Prospero et al., 2002), possibly indicating that either sediment 11 redistribution or non-eolian inputs may not be fully constrained in that region (Anderson et 12 al., 2014).

13 Significant underestimation of dust deposition by the model in Alaska is also suggested by the 14 observations. Note that dust sources in Alaska are glaciogenic, and in the model for the MH 15 we prescribed them; we allowed particular grid cells to emit dust with no constraints provided 16 by vegetation cover or geomorphic soil erodibility. The prescribed sources are the Matanuska 17 Valley, the Copper River Valley, and the belt in Central Alaska from Fairbanks to the West 18 coast, including the Yukon Valley. The total amount of dust that we allowed to be emitted 19 from Alaska as a whole is constrained by the fact that larger emissions would result in a 20 prevalence of Alaskan dust in Greenland in the model, which would not be consistent with 21 observations (e.g. Bory et al., 2003). Satellite imagery clearly shows that even in large dust 22 source areas, at a small spatial scale dust emanates from a constellation of localized hotspots, 23 and then gets mixed downwind (e.g. Knippertz and Todd, 2012). Global scale ESMs such as 24 the CESM have a spatial resolution good enough to capture the process of large spatial extent, 25 but may be more sensitive to the exact localization of small dust hotspots when they are scattered over disparate valley settings, as in the case of Alaska. An insight from a slightly 26 different angle could be that it is still unclear to what extent the very large DMARs from 27 28 localized sources in low hotspot density regions such as Alaska are representative for large 29 scale dust emissions, as discussed in Sect. 4.5.

The temporal evolution of the global dust cycle (Fig. 17) shows a decreasing trend in dustiness from the Early to Mid-Holocene, with a minimum between 6 and 8 ka BP, and an increasing tendency in the Late Holocene, with the global dust load varying between 17.1 and

1 20.5 Tg, which corresponds to a difference of ~17%. For reference, dust load estimates with

2 the same model are 23.8 Tg for current climate, and 37.4 Tg for the LGM (Albani et al.,

3 2014). Similarly, global dust deposition estimates during the Holocene vary by $\sim 16\%$,

4 between $\sim 2,900$ Tg a⁻¹ (10 ka BP) and $\sim 2,400$ Tg a⁻¹ (8 ka BP) (Fig. 17).

Two distinct features characterize the spatial distribution of dust during the Holocene. First, 5 6 the Early to Mid-Holocene is characterized by enhanced dustiness in the Southern 7 Hemisphere compared to the Late Holocene. Second, there are shifts between the relative 8 importance of Asian versus North African sources. Even in the Late Holocene though, there 9 seems to be an imbalance towards Asian sources, compared to present day. This may be 10 related to the difficulties of constraining the model to the observations in general and for the 11 North African regions in particular, although the relative role of North Africa as a dust source 12 may have actually increased significantly since the pre-industrial period due to much 13 increased dustiness (Mulitza et al., 2010).

14 4.13 Particle size distributions

15 The organization of the available size distribution data into a common binning scheme not 16 only provides the tool to relate DMARs on a common size range, but also allows comparing 17 modelled and observed size distributions (e.g. Albani et al., 2014; Mahowald et al., 2014). In 18 Figure 18 we make this kind of comparison for the 6 ka BP time slice, which highlights how 19 the observed size distributions (blue solid lines) is coarser close to the source areas, and 20 becomes finer for more remote dust deposits such as marine sediments or ice core archives. 21 While significant uncertainties and biases may affect the different observations of size 22 distributions (e.g. Mahowald et al., 2014; Reid et al., 2003), this relation between dust particle 23 size and long-range transport is widely recognised (e.g. Lawrence and Neff, 2009; Pye, 1995). 24 Modelled size distributions (red dashed lines) in general capture this trend, with coarser size distributions simulated for terrestrial deposits compared to dust deposition further away from 25 26 the dust sources. Notable exceptions are the Antarctic ice core sites, which exhibit coarse 27 distributions in the model. This feature was already observed in previous studies, and 28 attributed mainly to biases in transport in the CAM4-BAM, that is used for this study as well 29 (Albani et al., 2014; Mahowald et al., 2014). 30 Focussing on terrestrial deposits, we can also see the gradual tendency for the observed and

31 modelled size distributions to shift towards finer distributions for larger distances from the

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1 sources. For instance Weinan lays farther away from the major dust sources in the Ordos,

2 Badain Juran, and Tengger (Figure 17k), and shows the smallest relative contribution of dust

3 in the model's bin4 (5-10 µm) compared to the other sites in the CLP. Similarly, Zagoskin

4 Lake in Alaska lays farther away form the putative sources in the Yukon Valley, than OWR

5 does with respect to the Sand Dunes in Nebraska (Figure 171), and exhibits finer particle size

6 <u>distributions</u>.

7 The temporal variability in dust size distributions during the Holocene is very limited both in
8 the observations and the model (not shown).

9

10 **5 Conclusions**

Here we present the first study using an innovative approach to organize a paleodust compilation for the Holocene from different sedimentary archives, by collecting and evaluating dust records that allow the reconstruction of time series of eolian mass accumulation rates with size information, with relevance for medium to long range transport.

15 The resulting database has the following characteristics.

16 - It is concise and accessible. The main information for each site included in the compilation 17 is a time series including age (with uncertainty), dust MAR (with uncertainty), and size distribution (where available) standardized by the use of a common binning scheme. The data 18 19 are organized in ASCII tables with a coherent formatting, easily accessible by scripting or for 20 importing into spreadsheets. The data will be publicly accessible on the web and released with 21 this paper. We also provide a graphical overview that synthesizes "at-a-glance" the intrinsic 22 characteristics and uncertainties for all the different records included in the compilation. 23 Complementary to the data is a categorical attribution of the confidence level of each record, 24 in terms of providing a reliable quantitative DMAR time series of eolian dust relevant for 25 medium to long-range transport. Finally, we report detailed information of the dust size distributions when available. In particular when full size distributions were available (rather 26 27 than mineralogical size classes), we standardized them to a common binning scheme, to 28 facilitate comparability.

- It is detailed and flexible. On-going research often provides the opportunity of refined age
models for sedimentary records, so we left the compilation open for easy future updates. In
addition to the basic information mentioned above, we report the ancillary information

1 necessary to re-derive the dust MARs time series: the detailed depths and the relevant dust

variables, i.e. dust concentration or dust proxy concentration or dust fraction and bulk densityif applicable.

Its compilation was highly participatory. It results from an extensive collaboration among
scientists from the observational and modelling communities, which allowed more in depth
analysis beyond the original studies.

One merit of the database is also to document and archive the data, and the full size
distribution data in particular, which would otherwise risk being lost. In most cases only one
metric, typically the median, is reported in papers, and in fact some of the size distributions
that were once available were not retrievable already for this paper.

We focused on dust variability during the Holocene, with emphasis on the MH as a key PMIP scenario, and also in relation to the large amount of variability that affected the present world's largest dust source, North Africa, with the termination of the African Humid Period (deMenocal et al., 2000; McGee et al., 2013).

15 An integrated approach of merging data and modelling with the CESM allowed a spatially 16 consistent reconstruction of the global dust cycle and its variability throughout the Holocene. 17 Our simulations indicate that the global dust load showed significant variability ranging 18 between 172 and 208 Tg, with a minimum during the Early to Mid-Holocene. The 19 model/data compilation is likely to be useful to both dust and ocean biogeochemical 20 modellers, who may use iron and mineral dust as a ballast input to their model (e.g. Moore et 21 al., 2006), or for observational studies to allow them to put their cores into the context of 22 existing estimates of depositional fluxes (e.g. Winckler et al., 2008).

23 In addition we report on two relevant aspects that emerged from this work.

24 First, we showed how the dust size distribution of dust is intrinsically related to the DMAR:

25 ignoring this tight coupling would cause a misleading interpretation of the dust cycle, not only

26 for modelling studies but also in a broader sense.

27 Second, comparing DMARs within a consistent size range allows for a consistent analysis of

28 the spatial features of the global dust cycle, which are not deducible by the simple analysis of

29 relative timing and amplitude of the variations among different paleodust reconstructions.

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- 1 Our analysis shows that a knowledge gap in understanding relevant features of the global dust
- 2 cycle still exists, in particular for key regions such as North African and Asian dust sources,

3 where quantitative information on the dust cycle is limited or not fully consistent.

- 4 In our representation of the loess/paleosol data, we depict them as DMAR time series, which
- 5 is a rather innovative approach introduced in previous compilation efforts (Kohfeld and
- 6 Harrison, 2003), as well as in a few observational studies (e.g., Muhs et al., 2013a; Stevens
- 7 and Lu, 2009), but which is not widespread in the loess community at large. We tentatively
- 8 used an approach that privileges pairing of the time series with the soil sub-units stratigraphy.

9 Future work will be needed to better asses this, as well as alternative approaches.

10 The possibility of comparing not only the size range but also the size distributions of dust

11 particles offers additional tools to understand the spatial evolution of the dust cycle (as well as

12 its temporal variability in principle). At a given climate state, for instance, it allows relating

13 the records from different sites to the major dust sources.

14 The work presented in this paper provides the tools for relating DMARs and climate; future

work will need to place in context the dust records with the climate conditions of the different
regions, by comparing to other paleoclimate proxies.

We present a framework for future work on dust compilations, and although here we focused on the Holocene, future updates using this framework are intending to improve the compilation. In addition, the framework provided for this compilation can be extended to wider time periods in the future, for example, the full span of the last glacial cycle and the deglaciation, and the Late Holocene to present day, which would allow linking the past and the present dust cycle.

23

24 Appendix A: Description of the template database tables and site sheets

All records in this compilation include a basic piece of information: a time series of eolian DMAR, with 1σ uncertainty on both ages and DMARs (Supplementary Material), and a categorical attribution of the confidence level (Table 1).

28 Because each record is characterized by a different number of age points, a separate table is

29 associated to each record. In addition, a descriptive sheet is provided for each record, with a

- 30 graphical overview of the sampling of the profile and the time-dependent dust MAR with
- 31 uncertainties, as well as metadata. For sites where size information is available, an additional

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integrative table is provided, as well as a document with details about the re-binning
 procedure.

3 Each table in the database is a TAB-separated ASCII document, named after the site, as 4 reported in Table 1. The first four columns contain the basic information: Age (ka BP), Dust MAR (g m⁻² a⁻¹), Age error (ka), and Dust MAR error (g m⁻² a⁻¹). A second set of columns 5 includes data relative to the depth of the samples and their age: Depth top (cm), Depth bottom 6 7 (cm), Depth center (cm), Age top (ka BP), Age bottom (ka BP), Age center (ka BP). Finally, a 8 third set of columns contains information relevant for the dust MAR calculation: Sediment 9 Bulk MAR (g m⁻² a⁻¹), SBMAR relative error, Sediment Dry Bulk Density (g cm⁻³), SDBD relative error, SR (cm ka⁻¹), SR relative error, Eolian Contribution (fraction), EC (ppm), and 10 11 EC relative error. All entries are filled either with data or "NA"s. 12 The tables with size information are also TAB-separated ASCII documents. There are two 13 types of them, one with size classes, and one with the re-binned size distributions. The first four columns again contain the basic information: Age (ka BP), Dust MAR (g m⁻² a⁻¹), Age 14

error (ka), and Dust MAR error (g m⁻² a⁻¹). The other columns contain either the size classes
as reported in the original work, or the binned data, with upper and lower limits indicated in
the first two rows of the table. The numbers represent the percentage contribution of each bin
to the total dust mass. "NAs" indicate no data for bins outside the original measurements size
range.

The descriptive sheet is composed of three panels. The upper one shows the dust MAR in function of depth, and highlights (grey shading) the sampling stratigraphy. The central panel shows the dust MAR time series, with relative uncertainties. The bottom panel contains a concise summary about the sampling, and methods used to determine the ages, age model, SBMAR and EC (with relative uncertainties), and size.

25 For the records with size distributions associated, an additional PDF document is provided, showing the fitting procedure for each site: original (black) and new (red) cumulative 26 27 distributions, fitting spline (green), original (black) and new (red) mass-size distribution 28 (scaled). In addition, several percentiles across the size spectrum are compared for the original 29 and re-binned distributions. For the overall record from one site, two summary metrics are produced, which synthesize the overall fit to the original data: the Pearson's correlation 30 coefficient of the 5th, 25th, 50th, 90th, and 95th percentiles, and the mean normalized bias 31 32 (MNB):

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$$1 \qquad MNB = \frac{1}{n_{obs} * n_i} \sum_{obs} \sum_i \frac{(x_{obs,i}) - (y_{obs,i})}{x_{obs,i}} \tag{A1}$$

where n_{obs} is the number of samples for a site, n_i is the number of percentiles included in the
calculation (here five of them), x_{obs,i} is the original ith percentile for a given sample, and y_{obs,i}
is the corresponding new binning percentile. In this context, the MNB is a metric of the
average over- or under-estimation of the "coarseness" of the re-binned size distributions
compared to the original observations.

7

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1 Table 1. List of the records included in this compilation, with their exact location 2 (coordinates) and geographical localization (0 = Alaska, 1 = Greenland, 2 = North Africa and3 North Atlantic, 3 = Arabian Sea, 4 = North America, 5 = East Asia and North Pacific, 6 =4 Equatorial Pacific, 7 = South Atlantic, 8 = Antarctica, 9 = Australia), and the type of natural 5 archive. We also report the availability of size distributions or size classes ("yes" if included in the database), and the details of the estimation of the fine (< 10 μ m) fraction. Reference to 6 7 the original studies is provided in the second column from the right. The rightmost column 8 reports the details of how the percentage of $DMAR < 10 \mu m$ was calculated, based on either 9 the data reported in the database (see also Section 3.5), personal communications from the 10 authors of the original studies, or informed assumptions based on nearby observations as 11 described in Albani et al. (2014).

Site Longitud Latitud Are Archiv Confiden Size Reference Eolian dust MAR % e (deg. E) e (deg. a ce level distributio <10 um e N) ns 01 classes EDC 123.35 -75.1 8 ice high yes Delmonte et al., 2004 From size distributions core (Supplementary material) Vostok-BH7 106.8 -78.47 Delmonte et al., 2004 From size distributions 8 ice high ves (Supplementary core material) GISP2 322.37 72.58 1 ice medium no Mayewski et al., Assume 100% 1997 core (Steffensen, 1997; Albani et al., 2014) EN06601-339 502 4 9 1 8 2 marine high François et al., 1990 Assume 50% no 0038PG core (Ratmever et al 1999) Albani et al., 2014) EN06601-339 375 4 2 3 3 2 Francois et al 1990 50% marine high no Assume 0021PG (Ratmeyer et al., 1999; core Albani et al., 2014) EN06601-340.238 François et al., 1990 50% 2.46 2 marine high no Assume 0029PG (Ratmeyer et al., 1999; core Albani et al., 2014) OC437-07-349.37 30.88 2 marine medium yes McGee et al., 2013 From size distributions GC27 (Supplementary core material)

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OC437-07- GC37	344.882	26.816	2	marine core	high	yes	McGee et al., 2013	From size distributions (Supplementary material)		
OC437-07- GC49	342.146	23.206	2	marine core	high	yes	McGee et al., 2013	From size distributions (Supplementary material)		
OC437-07- GC66	342.14	19.944	2	marine core	medium	yes	McGee et al., 2013	From size distributions (Supplementary material)		
OC437-07- GC68	342.718	19.363	2	marine core	high	yes	McGee et al., 2013	From size distributions (Supplementary material)		
RC24-12	348.583	-3.01	2	marine core	high	no	Bradtmiller et al., 2006	Assume 50% (Ratmeyer et al., 1999; Albani et al., 2014)		
RC24-07	348.083	-1.333	2	marine core	high	no	Bradtmiller et al., 2006	Assume 50% (Ratmeyer et al., 1999; Albani et al., 2014)		
RC24-01	346.35	0.55	2	marine core	high	no	Bradtmiller et al., 2006	Assume 50% (Ratmeyer et al., 1999; Albani et al., 2014)		
V22-182	342.73	-0.53	2	marine core	high	no	Bradtmiller et al., 2006	Assume 50% (Ratmeyer et al., 1999; Albani et al., 2014)		
V30-40	336.85	-0.2	2	marine core	high	no	Bradtmiller et al., 2006	Assume 50% (Ratmeyer et al., 1999; Albani et al., 2014)		
PS2498-1	345.18	-44.25	7	marine core	medium	no	Anderson et al., 2014	Assume 100%		
RC27-42	59.8	16.5	3	marine core	high	no	Pourmand et al., 2007	Assume 60% (Clemens et al., 1998; Clemens and Prell, 1990; Albani et al., 2014)		
93KL	64.22	23.58	3	marine core	medium	no	Pourmand et al., 2004	Assume 60% (Clemens et al., 1998; Clemens and Prell, 1990; Albani et al., 2014)		
ODP138-	249	-3	6	marine	medium	no	McGee et al., 2007	Assume 100%		

848B-1H-1				core				
ODP138- 849A-1H-1	249	0	6	marine core	medium	no	McGee et al., 2007	Assume 100%
ODP138- 850A-1H-1	249	1	6	marine core	medium	no	McGee et al., 2007	Assume 100%
ODP138- 851E-1H-1	249	3	6	marine core	medium	no	McGee et al., 2007	Assume 100%
ODP138- 852A-1H-1	250	5	6	marine core	medium	no	McGee et al., 2007	Assume 100%
ODP138- 853B-1H-1	250	7	6	marine core	medium	no	McGee et al., 2007	Assume 100%
TT013-PC72	220	0	6	marine core	high	no	Anderson et al., 2006	Assume 100%
TT013- MC27	220	-3	6	marine core	high	no	Anderson et al., 2006	Assume 100%
TT013- MC69	220	2	6	marine core	high	no	Anderson et al., 2006	Assume 100%
TT013- MC97	220	0	6	marine core	high	no	Anderson et al., 2006	Assume 100%
TT013- MC19	220	-1.8	6	marine core	high	no	Anderson et al., 2006	Assume 100%
V28-203	180.58	0.95	6	marine core	high	no	Bradtmiller et al., 2007	Assume 100%
V21-146	163	38	5	marine core	medium	yes	Hovan et al., 1991	From size distributions (Supplementary material)
SO-14-08-05	118.38	-16.35	9	marine core	medium	no	Hesse and McTainsh, 2003; Fitzsimmons et al., 2013	Assume 100%
E26.1	168.33	-40.28	9	marine core	medium	yes	Hesse, 1994; Fitzsimmons et al., 2013	From size distributions (Supplementary material)
Zagoskin_La ke	197.9	63	0	lake	medium	yes	Muhs et al., 2003b	From size classes (Supplementary material): clay% + 1/4

silt%

Chitina	215.62	61.54	0	loess / paleos ol	medium	yes	Muhs et al., 2013	From size classes (Supplementary material): clay% + 1/4 silt%
Luochuan	109.42	35.75	5	loess / paleos ol	medium	yes	Lu et al., 2013	From size distributions (H. Lu, personal comm.)
Jiuzhoutai	103.75	36.07	5	loess / paleos ol	medium	no	Kohfeld and Harrison, 2003	Assume 23% (Maher et al., 2010)
Duowa	102.63	35.65	5	loess / paleos ol	medium	no	Roberts et al., 2001	Assume 42%: clay% + 1/4 silt%
Beiguoyuan	107.28	36.62	5	loess / paleos ol	medium	yes	Stevens and Lu, 2009	From size distributions (Supplementary material)
Xifeng	107.72	35.53	5	loess / paleos ol	medium	yes	Stevens and Lu, 2009	From size distributions (Supplementary material)
<u>Jingyuan</u>	<u>104.6</u>	<u>36.35</u>	<u>5</u>	<u>loess /</u> paleos <u>ol</u>	<u>medium</u>	<u>yes</u>	Sun et al., 2012	From size distributions (Supplementary material)
Weinan	<u>109.58</u>	<u>34.43</u>	<u>5</u>	<u>loess /</u> paleos <u>ol</u>	<u>medium</u>	<u>yes</u>	Kang et al., 2013	From size distributions (Supplementary material)
OWR	258.58	40.5	4	loess / paleos ol	medium	yes	Miao et al., 2007	From size classes (Supplementary material): clay% + 1/4 silt%
LRC	259.81	41.48	4	loess / paleos ol	medium	yes	Miao et al., 2007	From size classes (Supplementary material): clay% + 1/4 silt%

1 Table 2. Dust source areas in the CESM model, and scale factors expressed as anomalies with

2 respect to a reference period, derived from the observations. The first column lists the model

3 dust source areas. In the second column are listed the geographical regions where

4 observations are clustered, which are used to scale the dust from the corresponding model

5 macro-areas. The reference periods are 4 ka BP for 2 ka BP, and 6ka BP for 8 and 10 ka BP.

Source area	Anomaly forcing regions	2 ka BP	4 ka BP	6 ka BP	8 ka BP	10 ka BP
	10810110		51	51		
Alaska	0	0.6224	1	1	1.0800	1.3381
North America	4, 6	0.8961	1	1	0.8810	0.9452
(Southwest)						
North America (Midwest)	4	1.0081	1	1	0.9929	0.9481
North Africa	2	1.3350	1	1	1.0030	1.5563
Central Asia	3, 1	0.9628	1	1	1.1448	1.1448
East Asia	5, 1	1,0257	1	1	1,0304	1,0720
South America (Northern regions)	7, 8, 6	0.7396	1	1	1.1093	1.3482
South America (Patagonia)	7, 8	0.9995	1	1	1.1358	1.2313
South Africa	7, 8, 9	0.9777	1	1	1.1898	1.2764
Australia	9, 8	0.9723	1	1	0.5183	1.4452

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Figure 1. Schematic representation of the process of calculation of eolian DMAR (Dust Mass
 Accumulation Rate), and its relation to the SR (Sedimentation Rate), DBD (Dry Bulk
 Density), SBMAR (Sediment Bulk MAR), and EC (Eolian Content). DMAR (on age scale) is
 the typical path for loess/paleosol records, whereas DMAR (chronology) indicates the final
 step of the workflow when EC is also measured.

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Figure 2. Example of different resolution of SBMAR and EC (Clemens and Prell, 1990).

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9 Figure 3. Conceptual plot of the evolution of dust deposition flux (DF) and size distribution
10 (% sand) as a function of distance from the source.

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Figure 4. Upper panel: subdivision of the globe in different areas, based on the spatial distribution of data in this compilation (0 = Alaska, 1 = Greenland, 2 = North Africa and North Atlantic, 3 = Arabian Sea, 4 = North America, 5 = East Asia and North Pacific, 6 = Equatorial Pacific, 7 = South Atlantic, 8 = Antarctica, 9 = Australia). Bottom panel: time series (at a 2 ka pace) of the dust deposition anomaly with respect to 6 ka BP for the different areas, as estimated from the observations. Color-coding of the different areas is coherent between upper and lower panel.

20 Figure 5. Overview of the data compilation. Central plot: global overview of the location of 21 the palodust records. Color indicates the confidence level (red = high confidence, blue = 22 medium confidence). Marker's shape indicates if size distributions / classes are available 23 (filled circles = yes, empty diamonds = no). Framing plots: time series of bulk dust MAR in the different areas, normalized to their Holocene (0-12 ka BP) average (red solid line for 24 25 reference, which represents the time span over which DMARs were averaged in the original 26 DIRTMAP: Kohfeld and Harrison, 2001). Black solid lines represent high confidence 27 records; gray lines identify medium confidence records. Records are plotted in the 0-22 ka BP interval, to allow a comparison with DIRTMAP3 (Maher et al., 2010) data (as reported in 28 29 Albani et al., 2014), represented by their glacial/interglacial ratio (green solid circles). 30 Vertical color shading bands highlight the last millennium (pink), the MH (5-7 ka BP, salmon), and Last Glacial Maximum (18-22 ka BP, light blue). 31

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1	Figure 6. Detailed view of the dust records in the North Africa / North Atlantic region. Upper
2	panel: geographical location of the paleodust records. Bottom panel: time series of the bulk
3	(dotted lines) and "fine" i.e. $\leq 10~\mu m$ (solid lines) dust MARs. Color-coding is consistent
4	between upper and lower panel. Vertical grey solid lined mark the sub-periods within the
5	Holocene as described in Section 3.4 with a pace of 2 ka. <u>Please refer to the descriptive sheets</u>
6	in the Supplement for a graphical display of the uncertainties for each record.
7	
8	Figure 7. Same as Figure 6, for the Arabian Sea region.
9	
10	Figure 8. Same as Figure 6, for the North American region.
11	
12	Figure 9. Same as Figure 6, for Alaska.
13	
14	Figure 10. Same as Figure 6, for East Asia and the North Pacific Ocean.
15	
16	Figure 11. Same as Figure 6, for Greenland.
17	
18	Figure 12. Same as Figure 6, for the Equatorial Pacific.
19	
20	Figure 13. Same as Figure 6, the Australian region.
21	
22	Figure 14. Same as Figure 6, for the South Atlantic Ocean.
23	
24	Figure 15. Same as Figure 6, for Antarctica.
25	
26	Figure 16. Comparison of simulated dust deposition $(g m^{-2} a^{-1})$ for the 6 ka BP case, compared
27	to observational estimates of the fine (< 10 μ m) eolian Mass Accumulation Rate for the period
28	5-7 ka BP. (top) Observations; (middle) model; (bottom) model versus observations
29	scatterplot. Horizontal bars represent the variability of observational data averaged within the
30	5-7 ka BP time lapse (1 sigma). Locations of observational sites are clustered in the
21	anothermulate based on their accommunical location, on indicated by the color adding. In the

scatterplots based on their geographical location, as indicated by the color-coding. In the
bottom scatterplot, squares indicate high confidence level, diamonds represent medium
confidence level.

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Figure 17. Dust deposition flux (g m⁻² a⁻¹) from the CESM during the Holocene snapshots at
2, 4, 6, 8, and 10 ka BP, based on spatially variable emissions constrained by the
observational Mass Accumulation Rates. Black circles mark the locations of the observational
records in this compilation.

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7 Figure 18. Comparison of modelled and observed particle size distributions for the 6 ka BP time slice. Panels a-j show the modelled size distribution (red dashed line) and the observed 8 9 particle size distribution (blue solid line). The normalized observational size data from the re-10 binned distributions were first averaged over the 5-7 ka BP interval; then the size distribution 11 data were aggregated, in order to match the model dimensional bins (highlighted by the 12 horizontal grey bars). Both the modelled and observed size distributions are normalized over 13 the model size range, i.e. over the four size bins. Panels k and l show the relative geographical 14 position of terrestrial records (red empty squares) and the model dust sources (filled grey 15 squares) for East Asia and North America, respectively. Light grey squares indicate modelled 16 dust mobilization flux > 0, and dark grey squares denote the major dust sources, i.e. mobilization flux $> 200 \text{ g m}^{-2} \text{ a}^{-1}$. 17

