

## Response to Reviewers

We thank the reviewers for their comments, which helped to improve the manuscript. We answer each comment and how we changed the MS accordingly. Please note, that the values of the new global calculations changed slightly because we ran the re-calculations with a 20km grid resolution and modified the global map from shape to grid format to allow for faster calculations.

We would kindly ask to delete “Short Communication” in the title.

Response to Referee 1:

<p>1) it is somehow surprising that the weathering of old basaltic surfaces does not depend on the local runoff value. The “expected” weathering law only depends on temperature, based on the plot displayed on fig 1b. Would it be useful to plot the CO<sub>2</sub> consumption by the weathering of old basaltic outcrop as a function of runoff ?</p>	<p>These patterns were already analyzed in Li et al. (2016). The temperature normalization is used here, because of the robustness of the pattern.</p> <p>The correlation between runoff and alkalinity flux rates is weaker than using temperature. This still holds if including the uncertainties of the data (with Monte Carlo type analysis, <math>R^2=0.96</math>, <math>p&lt;10^{-3}</math>, using a linear regression for observed alkalinity flux rates and calculated alkalinity flux rates using the new temperature scaling law). Considering the Monte Carlo type application, the correlation between runoff and alkalinity flux rates becomes even weaker, if compared to a regression analysis using only the single data points.</p> <p>It is not concluded that runoff plays not a significant role for determining basalt weathering rates. However, the correlation between runoff and alkalinity flux rates is not significant any longer when Inactive Volcanic fields (IVFs) and Active Volcanic Fields (AVFs) are considered separately (new Figure 2c) (Li et al., 2016).</p> <p>We included a figure showing the runoff/alkalinity relationship of old volcanic fields, now in the main text (Fig.2c).</p>
<p>2) The authors implicitly assume that the young volcanic area are basaltic. This is not always the case (see for instance Rad et al., 2013, J South Am Earth Sci) . There is a possibility of bias in the present database: the old surfaces being basaltic, while the young volcanic areas can be dominated or affected by an andesitic lithology.</p>	<p>We consider now only mapped areas in catchments that were clearly described as “basaltic” or dominated by basalt. We included this part in the discussion in line 185-191: “The lithologies, predominantly described as basaltic in the global map, might introduce an additional bias to the global calculations because heterogeneities in the lithology cannot be excluded. Two active</p>

	volcanic fields (Sao Miguel and Tianchi Lake) were excluded from the calculation of the scaling law function because available catchments with alkalinity data hold largely lithologies of trachytic composition. Nevertheless, their data points (Fig. 1c) seem to show the same weathering behavior.”
3) There is no discussion about the contribution of ash weathering to the alkalinity flux. Ashes released by active volcanoes may represent an important contributor to the CO <sub>2</sub> consumption by weathering given their high reactive surfaces and their content in glass (see for instance Sowards et al., 2018, Geosphere). I think this should discuss.	<p>We agree with this and included it in line 140-143 in the discussion:</p> <p>“Volcanic ashes and ejecta might contribute to elevated weathering fluxes because of a relatively high content of glass. Glass dissolution rates are relatively high compared to mineral dissolution rates in general, but base cation content release varies dependent on the Si:O ratio (Wolff-Boenisch et al., 2006).”</p> <p>We agree that volcanic ashes provide fresh weatherable material on top of the basaltic rocks.</p>

Response to Referee 2 (R. Emberson):

<p>1. I think it is imperative that the authors provide uncertainty / error estimates on all of their results, and describe how these estimates were made (probably in the supplemental material). Without estimates of uncertainty, the reader is unable to assess the significance of the results. Even if the uncertainty on the measurements has already been made in the prior work (Li et al. 2016), I feel it’s essential that the authors also show these estimates (and additionally explain again how these estimates were derived – see comment number 5 for further discussion). Specifically, I’d like to see uncertainty estimates attached to all plotted points in the figures, in equations 2 and 4, and in the estimates in Table 1.</p>	<p>We agree with this and included the uncertainty estimates by Li et al. (2016) in all figures where it was possible. It is not possible to give a suitable uncertainty for the mapping accuracy of the Holocene fraction due to the nature of the data.</p> <p>We calculated the mean residual standard deviation of the newly derived function using a Monte Carlo method and show the standard deviation in eq.2 and 5. The calculation of the mean residual standard deviation is described in the supplemental material.</p> <p>The values of the global calculation of the CO<sub>2</sub> consumption fluxes were calculated by applying each 10,000 (Levenberg-Marquardt-algorithm) non-linear correlations to a new global basalt map and global datasets of temperature and runoff (Fekete et al., 2002; Hijmans et al., 2005). From this we calculated the mean global values of the fluxes, their standard deviations and percentiles (25, 75) (Supplemental Material F in the supplemental material).</p>
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<p>2. The different behavior of IVF and AVF areas based on reactivity is a novel way to explore the basaltic weathering regime, but perhaps it would be useful for some readers if the authors also explained this in the context of supply-limited or kinetic-limited weathering (e.g. Ferrier et al. 2016 <i>Geochemistry Geophysics Geosystems</i>). The IVF behavior shown in Fig 1b certainly looks like classic kinetic-limited weathering, but as the authors point out in the AVF this relationship no longer holds. I would suggest plotting the AVF data in the same way as the IVF data in Fig 1b. to see if there is a similar kinetically-limited relationship; if so, this could indicate that the boundary conditions for the kinetically limited system differ (e.g. due to less clay precipitation in younger lava bodies), whereas if such a kinetically-limited behavior is not evident (or the residuals are very large) then perhaps other effects (e.g. magmatic CO<sub>2</sub> degassing) may be relevant. Showing the reader these data could help demonstrate the effects seen, as well as making the relationships easier to understand for geochemists who work in the kinetic/supply limited paradigm.</p>	<p>The new Fig.2d shows the IVFs and AVFs combined in a temperature/alkalinity plot. The scattering in the plot shows that there is no classic kinetic-limited weathering visible for AVF, while, as described, the IVF follow an apparent temperature dependency. Taking into account that the four AVFs Mt. Etna, Mt. Cameroon, Virunga and La Reunion have the highest Holocene fractions (96.6%-27.7%) and all the other AVFs, which have less than 15% Holocene coverage, supports the argument that elevated geothermal fluxes, magmatic CO<sub>2</sub> and fresh material supplied on top of the classical critical zone contribute to enhanced alkalinity fluxes of these volcanic fields, as supported by referenced studies.</p> <p>Therefore, the classical concept of supply versus kinetic limitation in tectonic context and referring to a “shallow” critical zone as discussed in Ferrier et al. (2016) does not work here. We included some discussion for this before pointing out the relevance of intra-volcanic weathering (not the shallow critical zone), and the supply of fresh material due to volcanic activity.</p> <p>We include the reference of Ferrier et al. (2016) now and added a discussion part as pointed out by the reviewer.</p>
<p>3. The authors explain that they use the Holocene transition as a way to split the AVFs from the IVFs. This makes sense in terms of data availability, and I think it is an appropriate (if arbitrary) way to separate the data. However, the Holocene transition involved a series of global climatic changes, and I think it would be useful for the authors to explore whether this transition and the climatic changes associated with it could explain the differences observed in ‘reactivity’ between pre and post Holocene basaltic fields. I appreciate that the AVF and IVF areas both experience the same climate today, but I think it would be at least useful to discuss whether or not there could be a legacy effect in the IVF areas from weathering under a different climatic regime.</p>	<p>Our study is based on today’s climate conditions and it is unknown how different climate regimes in the past may have influenced the basalt weathering fluxes due to the lack of data on the distribution of young surface areas in the past. This point is difficult to address since we have only observation data of today and it is not possible for now to reconstruct active volcanic areas for the past in a global compilation. After times of globally warmer climate and higher erosion rates, the difference in reactivity between IVFs and AVFs might be reduced, but we lack information for this in our global compilation of data.</p> <p>Furthermore, we calculated global fluxes for the Mid-Holocene using Earth System model outputs, comparing the different alkalinity flux models. The calculations show that models, which consider only runoff as parameter provide elevated fluxes in the Mid-Holocene, of 8 %, due to slightly higher global runoff values (Bluth and Kump, 1994; Amiotte-Suchet and Probst, 1995), while models considering temperature and runoff provide lower values (-32 to 36%; (Dessert et al., 2003; Goll et al., 2014).</p>

	<p>Considering only temperature (new scaling equation developed for today's settings) results in only small differences in the global weathering fluxes of 10% compared to today, for areas with &gt; 74 mm/a runoff. These calculations are based on highly speculative assumptions, like geographical distribution of active volcanic areas are as today.</p> <p>Specifically, we do not know the Holocene-equivalent area if using mid-Holocene as reference point for example. Therefore, we are not sure, what the reviewer wants us to do here. We do not understand how a "legacy" effect can be identified based on the available data and how this could be quantified.</p> <p>Therefore, we would like to keep this calculation out of the manuscript.</p>
<p>4. It would be helpful to provide detail of the water chemistry measurements or data involved in this study in the supplemental material. From the information provided, it is not possible for the reader to tell how variable the HCO<sub>3</sub>-measurements made were, which makes it hard to judge the estimates of CO<sub>2</sub> sequestration. While I appreciate that a full assessment of the chemical composition of the rivers in question is significantly outside the scope of the study, it would be helpful for clarity if the authors discussed the following points, either in main or supplemental text: How variable is the HCO<sub>3</sub> in the water? Are the rivers supersaturated with respect to calcite? This is a fairly important issue, as there is potential for secondary carbonate precipitation to form in the rivers and soils of a catchment, which means the final estimates of HCO<sub>3</sub> flux may be biased. What are the concentration – discharge relationships for HCO<sub>3</sub> in these rivers? If concentration is relatively high even at high discharge (i.e. are the rivers near-chemostatic? - e.g. Godsey et al. 2009, Hydrological Processes) then the largest storm events have the greatest importance for HCO<sub>3</sub> flux – and as a result, changes in climate across the Holocene transition (e.g. different storm frequency) may be relevant for the findings (see point 3 above). As I say above, I fully appreciate that data on these points may be lacking, and certainly addressing these points in full is outside the scope of this study – I would just suggest</p>	<p>We tried to address this important point here and put some sentence into the supplement:</p> <p>For six areas water chemistry data are available (High Cascades, Japan, NE North America, SE Australia, South Africa and Tasmania), while for others either pH or major cations were not available.</p> <p>For these available data (103 catchments) the saturation index for calcite was calculated. In general, water samples are undersaturated with respect to calcite (77%). From the oversaturated samples 50% have values close to 0 with an saturation index SI &lt; 0.5. SI ~ 0.5 is the typical value for rivers in limestone areas (Romero-Mujalli et al., 2018). The other 12 values are between SI=0.5 and 0.9 and according water sample locations are mostly in dry areas of South Africa or Australia.</p> <p>In general, younger active areas have significant contributions of magmatic SO<sub>4</sub> or Cl, which shifts the saturation states normally further to lower, negative values.</p> <p>We cannot conclude from the river data what happens in the aquifer system but reference the study of Jacobson et al. (2015), which quantified the contribution of trace calcite dissolution from basalt using Ca-isotope data.</p>

<p>explaining how these issues may relate to your results, and what your assumptions are with regards to the river chemistry. This would really help the reader appreciate the results. A discussion of the assumptions could be incorporated into supplemental material.</p>	
<p>5. This study is a useful addition to the study of Li et al. (2016), from which much (if not all?) of the data seems to be drawn. While I appreciate that the prior work is a published study, I think it is important that the authors explain their methodology in this publication too. For example, it would be useful to explain how alkalinity calculations were made, and some of the assumptions associated with the chemistry data; it would also be useful to summarise all of the uncertainty estimates made in that prior study in this study (see point 1). I notice that the Li et al. 2016 paper is open-access (much appreciated!), so I can appreciate that some researchers may feel it is sufficient to just cite the methods in the prior work. My personal preference is for as much of the relevant methodology for a given study to be described in that study as possible, but I leave this to the editor's discretion in this case.</p>	<p>Li et al. (2016) calculated the alkalinity flux rates by multiplying the mean concentration of dissolved inorganic carbon (DIC) by the annual runoff. We included this explanation in the supplemental material. However, because the supplement is already very long, and the other data file is open access online, we would like to avoid replicating the data since the supplement is already rather long with over 40 pages, explaining the new data and calculations. Table S1 summarizes the hydrochemical data per volcanic area.</p>
<p>6. A small point, but one I feel worth mentioning – the HCO<sub>3</sub>/reactivity figures in the supplementary material (lines 188-189) are a really useful accompaniment to Figure 1 in the main text, and I would suggest incorporating at least some of them into the main text. It really helped me understand the importance of temperature and runoff, and I think they're important enough to include. Even if you choose not to do so, it would be useful to ensure that the symbology in the figures corresponds to one another (i.e. red-blue points in the main text, red-green points in the supplement). I'd suggest using red-blue as in the main text, to avoid any issues with red-green colour blind readers.</p>	<p>We incorporated two of the figures in the main text and changed the colors to blue and red (now new: Figure 2a to 2d compare the relationships).</p>

<p>7. It may be useful to compare these findings to those relating to weathering in aging glacial moraines, to help contextualize the importance of aging? Your results are really intriguing, and provide impetus for research questions focused on e.g. weathering in lava flows of known age (via e.g. cosmo dating) and comparing to glacial moraines would be direct comparison.</p>	<p>Indeed, we included a reference from Taylor and Blum (1995), discussing this in the introduction, lines 44-45.</p>
<p>8. In line 40, you refer to ‘geogenic nutrients’. I think this is a jargon term that many readers won’t understand; I’d suggest defining this term before you use it. Additionally, I think this statement needs a citation to support it.</p>	<p>This is a normal scientific use of common terms geo and genic. Geogenic means here derived from rock/lithosphere, comparable to the term anthropogenic.</p>
<p>9. In a couple of locations, you separate the arid and non-arid locations based on a rainfall total of &gt;74mm per year – why was this figure chosen? It would be helpful to have some explanation as to this number.</p>	<p>As we explained in the text before <math>74 \text{ mm a}^{-1}</math> was the lowest runoff value of a volcanic field in the dataset of Li et al. (2016). We included an explanation in line 109-114 and show in the summary table of model comparison that the cutoff is necessary to avoid overestimation due to the temperature scaling law used.</p>
<p>10. I would suggest arranging references in chronological order where there are multiple citations in parentheses, e.g. Line 43 and other locations.</p>	<p>We agree with this point.</p>
<p>Typographical / Syntax points In general I found the paper to be well written and easy to read. There are a handful of places where the language is somewhat idiosyncratic, and I’ve tried to make suggestions where possible. As a general point I would recommend using the active voice rather than passive voice to improve readability, but that’s probably a matter of personal preference.</p>	<p>Thank you very much.</p>
<p>Line 12: Consider being more specific than the word ‘information’ – perhaps describe which types of data you mean.</p>	<p>We rewrote this sentence.</p>
<p>Line 20: Instead of “from surface near material in the critical zone”, consider “from material in the shallow critical zone”</p>	<p>We agree and changed it correspondingly.</p>
<p>Line 22: Remove comma after suggests</p>	<p>We deleted the comma.</p>
<p>Line 23: “Active basalt areas” is jargon – I’d suggest it’s best to define what you mean in the main text</p>	<p>We deleted “active”.</p>

and avoid using jargon terms in the abstract	
Line 30: “Basalt areas, despite its limited...” should be “Basalt areas, despite their limited...”. Also remove the ‘the’ before CO2.	We agree and changed it.
Line 39-40: Consider changing “The role of basalt weathering in the carbon cycle and its feedback strength in the climate system depends, besides the release of ...” to “The importance of basalt weathering in the carbon cycle and the climate-weathering feedback loop depends in part on the release of geogenic nutrients but crucially on the amount of associated...”	We changed it to: “The role of basalt weathering in the carbon cycle and its feedback strength in the climate system depends, besides the release of geogenic nutrients, on the amount of associated CO <sub>2</sub> consumption and related alkalinity fluxes.”
Line 59: Replace “However, the here suggested aging effect” with “However, the effect of aging on weathering rates from a volcanic system discussed here has not been evaluated.”	We replaced this part.
Line 68: Consider replacing “the fraction of the Holocene area on the total studied area” with “the proportion of total area occupied by Holocene lavas”	We changed this part.
Line 92-93: The word order and verb agreement in this sentence is somewhat unclear – I haven’t suggested a revision since I don’t want to mess with the meaning, but I’d suggest revising it to clarify what you mean.	We modified this part.
Line 100: Replace “For allowing comparison with” with “to allow for comparison with”	We replaced this.
Line 103: Replace ‘reporting’ with e.g. ‘describing’	We agree and changed it.
Table 1: The first two columns in the table have no label? Please add a label to explain what these are.	We changed the labels of the table.
Line 152: Consider changing the phrase ‘time stamp’ – it isn’t necessarily clear what you mean.	We replaced it by “time period”.
Line 159: Replace ‘Results’ with “Our results”. Also change ‘considering’ to ‘exploring’	We changed this part.
Line 160: ‘Displacement’ – do you mean ‘emplacement’?	We replaced it.
Line 164: I would suggest rephrasing to remove the comma.	We modified this part.

Final paragraph: Consider re-stating your key finding in the final paragraph.	We rewrote this part.
Supplementary Material: Generally I found the supplementary material to be clear and helpful. Please ensure that uncertainty estimates are included where possible. I would also suggest checking with the editor as to whether the citations in the supplementary material will be indexed or not – it may be advisable to move them to the main text to ensure they get indexed.	Uncertainty estimates are now included in the supplemental material.

### References:

- Amiotte-Suchet, P., and Probst, J. L.: A global model for present-day atmospheric/soil CO<sub>2</sub> consumption by chemical erosion of continental rocks (GEM-CO<sub>2</sub>), *Tellus B*, 47, 273-280, 10.1034/j.1600-0889.47.issue1.23.x, 1995.
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- Dessert, C., Dupré, B., Gaillardet, J., François, L. M., and Allègre, C. J.: Basalt weathering laws and the impact of basalt weathering on the global carbon cycle, *Chemical Geology*, 202, 257-273, <http://dx.doi.org/10.1016/j.chemgeo.2002.10.001>, 2003.
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- Goll, D. S., Moosdorf, N., Hartmann, J., and Brovkin, V.: Climate-driven changes in chemical weathering and associated phosphorus release since 1850: Implications for the land carbon balance, *Geophysical Research Letters*, 41, 3553-3558, 10.1002/2014GL059471, 2014.
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- Romero-Mujalli, G., Hartmann, J., and Börker, J.: Temperature and CO<sub>2</sub> dependency of global carbonate weathering fluxes – Implications for future carbonate weathering research, *Chemical Geology*, <https://doi.org/10.1016/j.chemgeo.2018.08.010>, 2018.
- Taylor, A., and Blum, J. D.: Relation between soil age and silicate weathering rates determined from the chemical evolution of a glacial chronosequence, *Geology*, 23, 979-982, 1995.
- Wolff-Boenisch, D., Gislason, S. R., and Oelkers, E. H.: The effect of crystallinity on dissolution rates and CO<sub>2</sub> consumption capacity of silicates, *Geochimica et Cosmochimica Acta*, 70, 858-870, 2006.





1 **Short Communication: Aging of basalt volcanic systems and decreasing CO<sub>2</sub> consumption**  
2 **by weathering**

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9 **Abstract**

10 Basalt weathering is one of many relevant processes balancing the global carbon cycle via land-  
11 ocean alkalinity fluxes. The CO<sub>2</sub> consumption by weathering can be calculated using alkalinity  
12 and is often scaled with runoff and/or temperature. Here it is tested if ~~information on~~ the surface  
13 age distribution of a volcanic system derived by geological maps is a useful proxy for changes in  
14 alkalinity production with time.

15 A linear relationship between temperature normalized alkalinity fluxes and the Holocene area  
16 fraction of a volcanic field was identified, using information from 33 basalt volcanic fields, with  
17 an  $r^2=0.9493$ . This relationship is interpreted as an aging function and suggests that fluxes from  
18 Holocene areas are ~10 times higher than those from old inactive volcanic fields. However, the  
19 cause for the decrease with time is probably a combination of effects, including a decrease in  
20 alkalinity production from ~~surface near~~ material in the shallow critical zone as well as a decline in  
21 hydrothermal activity and magmatic CO<sub>2</sub> contribution. The addition of fresh reactive material on  
22 top of the critical zone has an effect in young active volcanic settings which should be accounted  
23 for, too.

24 A comparison with global models suggests; that global alkalinity fluxes considering Holocene  
25 ~~active~~ basalt areas are ~~~7060~~% higher than the average from these models imply. The contribution  
26 of Holocene areas to the global basalt alkalinity fluxes is today however only ~~~65~~%, because  
27 identified, mapped Holocene basalt areas cover only ~1% of the existing basalt areas. The large  
28 trap basalt proportion on the global basalt areas today reduces the relevance of the aging effect.  
29 However, the aging effect might be a relevant process during periods of globally, intensive  
30 volcanic activity, which remains to be tested.

31 **1. Introduction**

32 Basalt areas, despite ~~its~~their limited areal coverage, contribute significantly to ~~the~~ CO<sub>2</sub>  
33 sequestration by silicate rock weathering (~~Dessert et al., 2003~~; Gaillardet et al., 1999; Dessert et

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34 [al., 2003](#); Hartmann et al., 2009). The sensitivity of basalt weathering to climate change ([Coogan](#)  
35 [and Dosso, 2015](#); Dessert et al., 2001; Dessert et al., 2003; [Coogan and Dosso, 2015](#); Li et al.,  
36 2016) supports a negative weathering feedback in the carbon cycle that maintains the habitability  
37 of the Earth's surface over geological time scales ([Walker et al., 1981](#); Berner, [et al., 1983](#); Li and  
38 Elderfield, 2013; [Walker et al., 1981](#)). Changes in volcanic weathering fluxes due to emplacement  
39 of large volcanic provinces or shifts in the geographic distribution of volcanic fields associated  
40 with continental drift may have contributed to climate change in the past (Goddéris et al., 2003;  
41 [Schaller et al., 2012](#); Kent and Muttoni, 2013; [Schaller et al., 2012](#)).

42 The role of basalt weathering in the carbon cycle and its feedback strength in the climate system  
43 depends, besides the release of geogenic nutrients, on the amount of associated CO<sub>2</sub> consumption  
44 and related alkalinity fluxes. The factors that modulate these fluxes are a subject to uncertainty.  
45 Previous studies suggest that basalt weathering contributes 25–35% to the global silicate CO<sub>2</sub>  
46 consumption by weathering ([Dessert et al., 2003](#); Gaillardet et al., 1999; [Dessert et al., 2003](#);  
47 Hartmann et al., 2009). However, their estimations do not consider the potential aging of a  
48 weathering system: (e.g., [Taylor and Blum, 1995](#)). Young volcanic areas can show much higher  
49 weathering rates compared to older ones, as was shown for the Lesser Antilles, where a rapid decay  
50 of weathering rates within the first 0.5 Ma was observed (Rad et al., 2013). Such an aging effect  
51 of volcanic areas is difficult to parameterize for global basalt weathering fluxes, due to a lack of  
52 global compilations.

53 A practical approach to resolve this issue is to distinguish ~~older~~ and inactive volcanic fields  
54 (IVF) and active volcanic fields (AVF) (Li et al., 2016) and compare weathering fluxes with factors  
55 driving the weathering process, like land surface temperature or hydrological parameters. By  
56 compiling data from 37 basaltic fields globally, Li et al. (2016) showed that spatially explicit  
57 alkalinity fluxes (or CO<sub>2</sub> consumption rates) associated with basalt weathering correlate strongly  
58 with land surface temperature for IVFs, but not for AVFs. They suggested that previously observed  
59 correlations between weathering rates and runoff in global datasets originates partly from the  
60 coincidence of high weathering rates and high runoff of AVFs rather than a direct primary runoff  
61 control on the weathering rate. Many studied AVFs are located near the oceans and have an  
62 elevated topography, a combination, which can cause elevated runoff due to an orographic effect  
63 (Gaillardet et al., 2011). However, the ~~here suggested effect of~~ aging ~~effect~~ on weathering rates  
64 from a volcanic system [discussed here](#) has not been evaluated.

65 The age distribution of the surface area of a whole volcanic system might be used as a first order  
66 proxy to study the variability of weathering fluxes of AVFs. However, the exact surface age of  
67 volcanic areas is rarely mapped in detail, but Holocene areas are often reported in geological maps.  
68 Here, basalt alkalinity fluxes are related to the calculated Holocene areal proportion of volcanic  
69 fields at the catchment scale. For this, the concept of weathering reactivity is introduced, which is  
70 the relative alkalinity flux of AVFs to the alkalinity flux estimated for IVFs ~~by the function~~  
71 ~~identified in Li et al. (2016). This reactivity R is compared with the relative age distribution of~~  
72 ~~surface areas, using the fraction of the Holocene area on the total studied area.~~ [This reactivity R](#)

73 is compared with the relative age distribution of surface areas, using the proportion of total area  
74 occupied by Holocene lavas. From this comparison a function for the decay of alkalinity fluxes  
75 with increasing proportion of older land surface area is derived and discussed.

## 77 2. Methods

78 The volcanic fields used to establish the relationship between weathering reactivity and Holocene  
79 coverage are predominantly described as basalt areas (Li et al., 2016). Based on the availability of  
80 detailed geological maps, 33 volcanic provinces were selected, with 19 IVFs and 14 AVFs. A  
81 detailed description is given in the supplementary information. The 14 AVFs are geographically  
82 widespread and diverse (Fig. 1a). If the absolute age distribution of the volcanic rocks is available,  
83 the Holocene areas were mapped using the age range from 11.7 ka to present, according to the  
84 International Commission on Stratigraphy version 2017/02 (Cohen et al., 2013). If possible,  
85 coordinates of water sample locations were used to constrain catchment boundary to calculate the  
86 Holocene fraction for monitored areas. In some cases already existing alkalinity flux  
87 calculations were taken from Li et al. (2016). Detailed information on additional mapping and  
88 calculations for each system can be found in the supplementary information (SI).

~~The weathering reactivity (R) of each volcanic field is calculated by normalizing the observed  
alkalinity flux of the AVF ( $F_{observed}$ , in  $10^6 \text{ mol km}^{-2} \text{ a}^{-1}$ ) to that of the expected flux if the AVF  
would be an IVF ( $F_{expected}$ ) applying the previously identified weathering function for IVFs (Li et  
al., 2016):~~

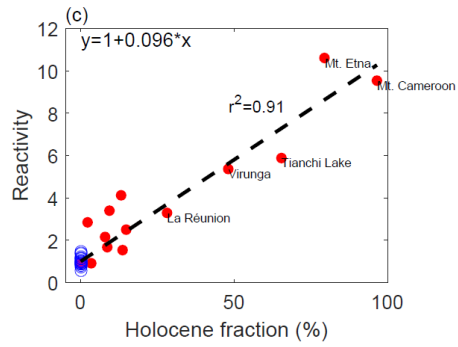
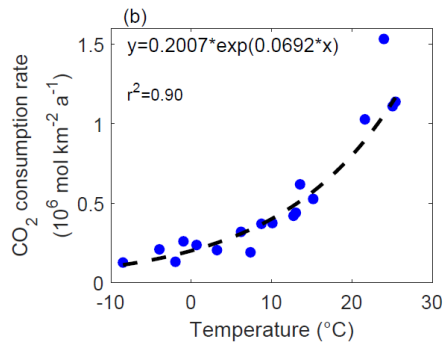
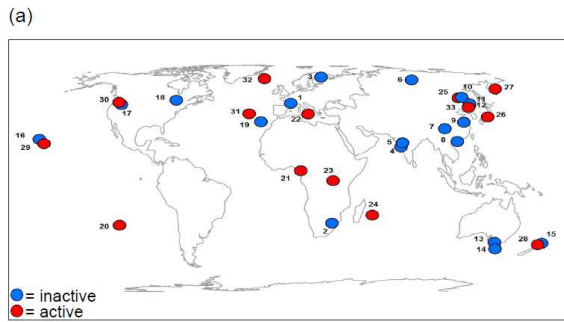
89 ~~$$R = F_{observed} / F_{expected} \quad \text{(eq. 1)}$$~~

~~The weathering reactivity (R) of each volcanic field is calculated by normalizing the observed  
alkalinity flux of the AVF ( $F_{calculated}$ , in  $10^6 \text{ mol km}^{-2} \text{ a}^{-1}$ ) to that of the expected flux if the AVF  
would be an IVF ( $F_{expected}$ ):~~

93 ~~$$R = \frac{F_{calculated}}{F_{expected}} \quad \text{(eq.1)}$$~~

95 where the expected alkalinity flux  $F_{expected}$  for IVFs is given by the function (Fig. 1b):

96 ~~$$F_{expected} (10^6 \text{ mol km}^{-2} \text{ a}^{-1}) = 0.2007 \times e^{(0.0692 \times T (^{\circ}\text{C}))} \quad \text{(eq. 2)}$$~~



97  $F_{expected} (10^6 \text{ mol km}^{-2} \text{ a}^{-1}) = 0.23 * e^{(0.06*T (^{\circ}C))}$ , standard deviation = 0.3 (eq.2)

98

99 The parameters of the equation were derived by using a Monte Carlo method, simulating 10,000  
 100 runs (for more information see Supplementary Information).

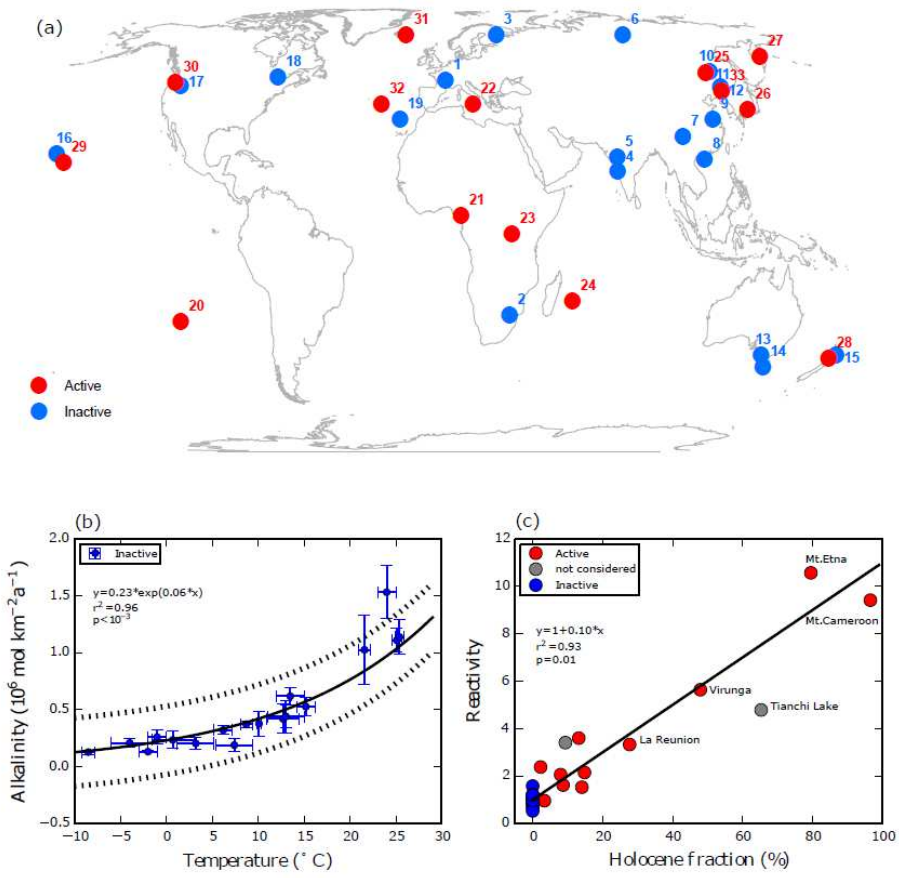


Figure 1. (a) The global map shows the locations of the Active Volcanic Fields (in red) and the Inactive Volcanic Fields (in blue) used in this study (1. Massif Central, 2. South Africa, 3. Karelia, 4. Coastal Deccan, 5. Interior Deccan, 6. Siberia Traps, 7. E' Mei, 8. Lei-Qiong, 9. Nanjing, 10. Xiaoxinganling, 11. Tumen River, 12. Mudan River, 13. SE Australia, 14. Tasmania, 15. North Island, NZ, 16. Kauai, Hawaii, 17. Columbia Plateau, 18. NE America, 19. Madeira Island, 20. Easter Island, 21. Mt. Cameroon, 22. Mt. Etna, 23. Virunga, 24. La Réunion, 25. Wudalianchi Lake, 26. Japan, 27. Kamchatka, 28. Taranaki, 29. Big Island, Hawaii, 30. High Cascades, 31. São Miguel, 32. Iceland, 33. Tianchi Lake). (b) The exponential relationship between alkalinity flux rates and the land surface temperature for IVFs (Li et al., 2016). (c) The relationship between the Holocene area fraction of a volcanic field and the weathering reactivity  $R$  ( $r^2=0.91$ )-Iceland.

32.Sao Miguel, 33.Tianchi Lake). (b) The exponential relationship between area specific alkalinity flux rates and the land surface temperature for IVFs. The dashed lines represent the range of the mean residual standard deviation of the function (see Supplementary Information). Note that  $r^2$  and the p-value were derived by a linear regression of calculated alkalinity flux rates vs. estimated alkalinity flux rates using the new scaling law. (c) The relationship between the Holocene area fraction of the used watersheds from the volcanic fields and the weathering reactivity R. Note that “Tianchi Lake” and “Sao Miguel” are excluded from the calculation of the regression line because the applied catchments were not dominated by basalt but by trachytic volcanic rock types. Both data points still seem to follow the identified regression trend line for AVFs.  $R^2$  and p-value were calculated by a linear regression of the Holocene fraction vs. reactivity for all AVFs.

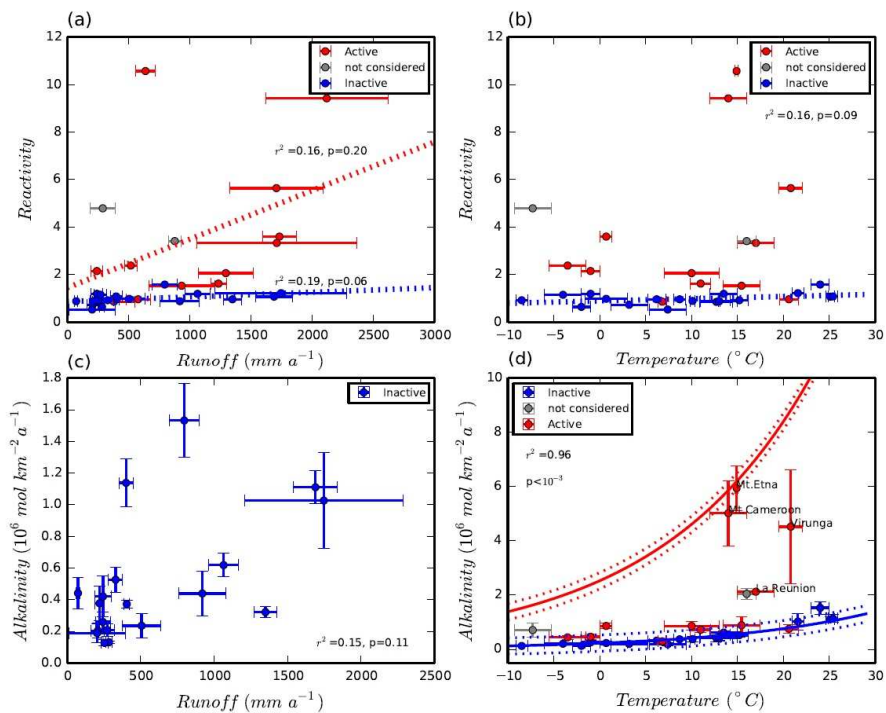
101 IVFs range group around a reactivity  $R=1$  in Fig. 1c, while having a Holocene fraction of zero (Fig  
 102 1e). The reactivity R (eq. 1) of an AVF can be estimated by the Holocene area fraction as implied  
 103 by the good significant linear correlation identified in Fig. 1c. The virtual theoretical reactivity for  
 104 a 100% Holocene area can be estimated by rearranging eq. 1 by the equation given by  
 105 Fig. 1c substituting y and x and setting the area fraction H to 100%:-

$$106 \frac{F_{Holocene}}{R_{100\% Holocene}} = \frac{1 + 0.10 * H}{11} = \frac{R_{100\%}}{11} \quad (eq.3)$$

109 And with this the flux from a young system of only Holocene  $* F_{expected}$   
 110 (eq. 3)age:

$$112 F_{Holocene} = R_{100\% Holocene} * F_{expected} \quad (eq.4)$$

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115

Figure 2(a) shows the runoff-reactivity relationship of all studied volcanic fields. The blue regression line (IVFs) suggests almost no correlation of reactivity with runoff, also for the AVFs no significant correlation is identified (coefficient of determination  $r^2=0.16$  ( $p=0.20$ )) (b) Reactivity versus temperature suggests no bias of reactivity due to a temperature effect for AVFs, as reactivity is based on a temperature normalized parameterization. For the IVFs ( $r^2=0.16$ ,  $p=0.09$ ) no bias with temperature can be identified due to the good correlation of alkalinity fluxes with land surface temperature (blue line) (c) Runoff versus alkalinity flux rates for inactive volcanic fields shows no significant correlation ( $r^2=0.15$ ,  $p=0.11$ ) (d) Temperature versus alkalinity flux rates for all volcanic fields of the study. The blue line represents the new scaling law for IVFs and the red line the new scaling law for AVFs with 100% Holocene area coverage (calculated deviation is represented by dashed lines).  $R^2$  and  $p$ -value are derived by a linear regression of calculated alkalinity flux rates vs. observed alkalinity flux rates for all volcanic fields.

116

117 Global alkalinity fluxes from basalt areas were calculated by using equation 2 for older areas  
 118 than of Holocene age and equation 2 and 4 for mapped Holocene areas equation 3. The model.



119 These equations are (eq. 2 and following, using information based on eq. 2) were calibrated for areas  
120 with a runoff > 74 mm a<sup>-1</sup> (lowest runoff value in data compilation of Li et al. (2016) and therefore  
121 limit of the model setup) to avoid too high alkalinity fluxes from drier areas with high temperature  
122 (e.g., the Sahara Desert), assuming that neglecting fluxes from areas with lower runoff are not  
123 biasing the comparison (Tab. S1, supplementary information). In this case an overestimation is  
124 avoided. For the global calculation of CO<sub>2</sub> consumption by the new scaling law, a Monte Carlo  
125 method simulating 10,000 runs was applied (see supplement information).

126 Results are compared with four previous global empirical alkalinity flux models (Bluth and Kump,  
127 1994; Amiotte-Suchet and Probst, 1995; Dessert et al., 2003; Goll et al., 2014; Suchet and Probst,  
128 1995). Alkalinity fluxes were translated into CO<sub>2</sub> consumption to allow for ~~allowing~~ comparison  
129 with previous literature. For all models the same data input was used: a newly compiled global  
130 basalt map (mostly derived by the basalt lithological layer from the GLiM, but enhanced by  
131 mapped Holocene areas (SI; Hartmann and Moosdorf, 2012), additional regional geological maps  
132 ~~reporting on describing~~ basalt areas and the maps of the volcanic fields used in this study, for  
133 detailed information see supplementary information), temperature (Hijmans et al., 2005) and  
134 runoff (Fekete et al., 2002).

### 136 **3. Results and Discussion**

137 Studied IVFs are characterized by a Holocene volcanic surface area of 0% with weathering  
138 reactivity R ranging between 0.65 and 1.56 (Fig. 1c). In contrast, AVFs show a large range of  
139 Holocene coverage, from 0.2% (High Cascades) to 96.6% (Mount Cameroon), and weathering  
140 reactivity between 0.9 (~~Easter Island,~~ High Cascades) and 10.6 (Mount Etna). The weathering  
141 reactivity correlates strongly with the percentage of Holocene area ( $r^2 = 0.9193$ ; Fig. 1c),  
142 suggesting Holocene surface area distribution to be a good predictor for the enhanced alkalinity  
143 fluxes from a volcanic system:

$$144 F_{alkalinity} = (1 + 0.096 \times H) \times 0.2007 \times e^{(0.0692 \times T)} [10^6 \text{ mol km}^{-2} \text{ a}^{-1}] \text{ (eq. 4)}$$

$$145 F_{alkalinity} = (1 + 0.10 * H) * 0.23 * e^{(0.06 * T)} [10^6 \text{ mol km}^{-2} \text{ a}^{-1}], \text{ standard deviation} = 0.3 \text{ (eq. 5)}$$

146 where H is the Holocene fraction of a volcanic system in % and T is land surface temperature in  
147 °C.

148 The Holocene fraction is not interpreted as the physical cause for elevated alkalinity fluxes.  
149 Instead, magmatic CO<sub>2</sub> contribution, geothermal-hydrothermal activity and the input of new  
150 volcanic material ~~to the surface (properties and “freshness” of the surface area for reaction) are~~  
151 ~~contributing to enhanced alkalinity fluxes, on top of the surface (properties and “freshness” of the~~  
152 ~~surface area for reaction) are contributing to enhanced alkalinity fluxes. Volcanic ashes and ejecta~~  
153 ~~might contribute to elevated weathering fluxes because of a relatively high content of glass. Glass~~

154 dissolution rates are relatively high compared to mineral dissolution rates in general, but base  
155 cation content release varies dependent on the Si:O ratio (Wolff-Boenisch et al., 2006).

156 The magmatic CO<sub>2</sub> contributions to alkalinity fluxes in young volcanic systems may be large in  
157 general, but data are scarce to evaluate the global relevance for AVFs. For the Lesser Antilles a  
158 magmatic contribution of 23 to 40% to the CO<sub>2</sub> consumed by weathering was identified (Rivé et  
159 al., 2013). High <sup>13</sup>C-DIC values suggest that magmatic CO<sub>2</sub> contributes significantly to the  
160 alkalinity fluxes from the Virunga system (Balagizi et al., 2015). The magmatic CO<sub>2</sub> contribution  
161 derived from volcanic calcite dissolution on Iceland was estimated to be about 10% of the  
162 alkalinity fluxes for the studied area (Jacobson et al., 2015). In case of the Etna 7% of the CO<sub>2</sub>  
163 emitted due to volcanic activity may be captured by weathering (Aiuppa et al., 2000). These  
164 examples suggest that significant amounts of magmatic carbon may be transferred to the ocean  
165 directly via intra-volcanic weathering from AVFs.

166 These examples show that in case of active volcanic fields the traditional view on kinetic versus  
167 supply limitation in the “shallow” critical zone in context of tectonic settings does not hold (e.g.,  
168 Ferrier et al., 2016). In contrary the supply of fresh material on top of the classical critical zone  
169 and the weathering from below the classical critical zone, suggests that these hot spots of silicate  
170 weathering in active volcanic areas, which contribute over proportionally to the global CO<sub>2</sub>  
171 consumption by silicate weathering, demand likely a different way of looking at it. The blue data  
172 points (IVF) in Figure 2d suggest a kinetic limited regime and follow a temperature dependency.  
173 The red points (AVF) however are located above the data points of IVF in general. Taking into  
174 account that the four AVFs Mt. Etna, Mt. Cameroon, Virunga and La Reunion have the highest  
175 Holocene fractions (96.6%-27.7%) and that the further AVFs, which have less than 15% Holocene  
176 coverage are located in general above the regression line for IVFs, supports the argument that a  
177 combination of elevated geothermal fluxes, magmatic CO<sub>2</sub> and fresh material supplied on top of  
178 the classical critical zone contribute to the observed elevated alkalinity fluxes for AVFs in general.

179 The calculated global basalt weathering alkalinity fluxes based on previous global models (Bluth  
180 and Kump, 1994; Amiotte-Suchet and Probst, 1995; Dessert et al., 2003; Goll et al., 2014; Suehet  
181 and Probst, 1995) give alkalinity fluxes ranging between 0.78 to 1.67 × 10<sup>12</sup> mol a<sup>-1</sup>. These values  
182 are different to previously published results based on the same models because a different  
183 geological map and climate data are used in this study. The new scaling law calculation based on  
184 the temperature dependence of weathering rate and the age dependence of weathering reactivity  
185 (eq. 4) results in higher global alkalinity fluxes of 21.9 × 10<sup>12</sup> mol a<sup>-1</sup> and 3.43 × 10<sup>12</sup> mol a<sup>-1</sup> for  
186 regions with > 74mm a<sup>-1</sup> runoff, and for all areas, respectively. The latter higher estimate is mainly  
187 due to the modeled contribution from dry and hot regions and shows that it is relevant to apply the  
188 runoff cut off.

189 Using the introduced new approach, considering the aging of a volcanic system, reveals that  
190 alkalinity fluxes from Holocene areas contribute today only 65% to the global basalt weathering  
191 alkalinity flux. This is because so far identified mapped Holocene volcanic areas cover only ~1%

192 of all basalt areas. This study did not include areas of less mafic volcanic areas, like andesites, or  
 193 middle American volcanics.

194 The Holocene area is probably underestimated due to information gaps in the reported age  
 195 information of the global map. The strong dependence of weathering reactivity on relative age of  
 196 the surface of a considered volcanic system suggests that it is relevant to know the global spatial  
 197 age distribution of volcanic areas in more detail. Therefore, a new global review of the age  
 198 distribution of basalt areas would be needed, which is beyond the scope of this study. The  
 199 lithologies, predominantly described as basaltic in the global map, might introduce an additional  
 200 bias to the global calculations because heterogeneities in the lithology cannot be excluded. Two  
 201 active volcanic fields (Sao Miguel and Tianchi Lake) were excluded from the calculation of the  
 202 scaling law function because available catchments with alkalinity data hold largely lithologies of  
 203 trachytic composition. Nevertheless, their data points (Fig. 1c) seem to show the same weathering  
 204 behavior.

205

*Table 1: Summary of CO<sub>2</sub> consumption rates for global basaltic areas applying different models and the new parameterization. For simplicity it was assumed that alkalinity fluxes equal CO<sub>2</sub> consumption. The percentiles of the values of the global calculation by the new scaling law (Monte Carlo method) can be found in the supplement information. The standard deviation is represented by “New scaling law – std” as described in the SI.*

206

		Global CO <sub>2</sub> consumption rate (10 <sup>9</sup> mol/a) for limited area in comparison (only areas with > 74mm/a runoff)	Global CO <sub>2</sub> consumption rate (10 <sup>9</sup> mol/a)
Dessert et al. 2003	runoff, temperature	1567	1573
Amiotte-Suchet & Probst, 1995	runoff	838	843
Bluth & Kump, 1994	runoff	739	754
Goll et al., 2014	runoff, temperature	1471	1477
<u>New equation</u>	<u>temperature</u>	<u>1992</u>	<u>3431</u>

207

<u>Models for comparison</u>	<u>parameters</u>	<u>Global CO<sub>2</sub> consumption rate (10<sup>9</sup> mol/a) for limited area in comparison (only areas with &gt; 74mm/a runoff)</u>	<u>Global CO<sub>2</sub> consumption rate (10<sup>9</sup> mol/a)</u>
<u>Dessert et al. 2003</u>	<u>runoff, temperature</u>	<u>1669</u>	<u>1684</u>
<u>Amiotte-Suchet &amp; Probst, 1995</u>	<u>runoff</u>	<u>863</u>	<u>870</u>

<a href="#">Bluth &amp; Kump, 1994</a>	<a href="#">runoff</a>	<a href="#">746</a>	<a href="#">761</a>
<a href="#">Goll et al., 2014</a>	<a href="#">runoff, temperature</a>	<a href="#">1566</a>	<a href="#">1580</a>
<a href="#">New scaling law</a>	<a href="#">temperature</a>	<a href="#">1930</a>	<a href="#">3300</a>
<a href="#">New scaling law – std (SI)</a>		<a href="#">90</a>	<a href="#">200</a>

208

209 The applied time ~~stamp~~[period](#) of the Holocene boundary suggests that the aging of the “weathering  
210 motor” of a basaltic volcanic area, including internal weathering, with declining volcanic activity  
211 is rather rapid. This implies that peaks in global volcanic activity have probably a short but  
212 intensive effect on the CO<sub>2</sub> consumption. A pronounced effect on the global carbon cycle by  
213 shifting the global reactivity of volcanic areas may only be relevant for geological periods with  
214 significantly elevated production of new volcanic areas, accompanied by geothermal-  
215 hydrothermal activity and capture of magmatic CO<sub>2</sub> before its escape to the atmosphere.

216 Results may have relevance for the carbon cycle and climate studies ~~considering~~[exploring](#) the  
217 ~~displacement~~[emplacement](#) of large igneous provinces like the CAMP (Schaller et al., 2012) or the  
218 Deccan traps (~~Caldeira, 1990~~)([Caldeira and Rampino, 1990](#)) with production of large basaltic areas  
219 within a short time. However, the biological contribution to CO<sub>2</sub> drawdown, via [elevated](#)  
220 fertilization effects, e.g. P- or Si-release due to weathering and elevated CO<sub>2</sub> in the atmosphere  
221 ~~must also~~[should](#) be taken into account, [too](#).

222 Looking deeper into Earth’s history: variations in the solid Earth CO<sub>2</sub> degassing rate or changes in  
223 environmental conditions affecting the weathering intensity ([Teitler et al., 2014](#); Hartmann et al.,  
224 2017; ~~Teitler et al., 2014~~) may have caused different reactivity patterns in dependence of surface  
225 age, ~~if compared to the ones identified for the recent conditions~~ [as shown here](#).

226 In conclusion, a simple approach to detect an aging effect, using surface age as a proxy for several  
227 combined processes, was chosen due to availability of data. [It can be shown that there exists a](#)  
228 [linear relationship between temperature normalized alkalinity fluxes and the Holocene area](#)  
229 [fraction of a volcanic system](#). Nevertheless, the combined effect on elevated weathering reactivity  
230 due to magmatic CO<sub>2</sub> contribution, hydrothermal activity, production of fresh surface area for  
231 reaction, and hydrological factors of young volcanic systems remains to be disentangled, for single  
232 volcanic systems, as well as for the emplacement of larger, trap-style basalt areas.

233

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1 **Supplementary Materials for**

2 **Short Communication: Supplement Information**

3 **Aging of basalt volcanic systems and decreasing CO<sub>2</sub> consumption by**  
4 **weathering**

5  
6 ~~This supplementary material describes~~ [Janine Börker<sup>1</sup>, Jens Hartmann<sup>1</sup>, Gibran Romero-](#)  
7 [Mujalli<sup>1</sup>, Gaojun Li<sup>2</sup>](#)

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9 [Hamburg, Bundesstraße 55, 20146 Hamburg, Germany. E-Mail: \[janine.boerker@uni-\]\(mailto:janine.boerker@uni-hamburg.de\)](#)  
10 [hamburg.de; geo@hattes.de](mailto:geo@hattes.de)

11 <sup>2</sup>[MOE Key Laboratory of Surficial Geochemistry, Department of Earth Sciences, Nanjing](#)  
12 [University, 163 Xianlindadao, Nanjing 210023, China](#)

13  
14 ~~Supplemental Material A to G describe~~ available information on ~~the used~~ volcanic, basalt  
15 ~~dominated~~ areas and ~~the~~ calculation procedures. For the active volcanic fields (AVF) a  
16 detailed description of the calculation for the ~~fraction of the~~ Holocene area ~~fraction on the~~  
17 ~~total area~~ is given. Additionally, a description of the ~~newnewly introduced~~ alkalinity flux  
18 ~~equationscaling law~~ for young active basaltic areas and ~~an explanation~~ for the calculation  
19 of the global fluxes is provided.

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20 **Part One**

21 **Supplemental Material A: Summary of data compilation as applied in the main text**

*Table S1: Summary of the data of IVFs and AVFs used for this study.*

22 **Supplemental Material B: New data for active volcanic fields as described in Table**  
23 **S1, in addition to Li et al. (2016)**

24 **Supplemental Material C: Saturation state with respect to calcite**

25 **Supplemental Material D: Additional relations between Alkalinity, Reactivity and**  
26 **Holocene area fraction**

27 **Supplemental Material E: Estimation of the parameters for the new scaling law**

28 **Supplemental Material F: Global calculations CO<sub>2</sub> consumption by basalt**  
29 **weathering**

30 **Supplemental Material G: Enhanced global basalt map beyond GLIM**

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8 **Supplemental Material A: Summary of data compilation as applied in the main text**

*Table S1: Summary of the data of Inactive Volcanic Fields (IVFs) and Active Volcanic Fields (AVFs) used for the analysis in the main text (Li et al., 2016), as well as calculated Holocene fraction and Reactivity R.*

No	Name	Volcanic activity	T (°C)	$\sigma_T$	Runoff (mm/yr)	$\sigma_{Runoff}$	HC $CO_3^{2-}$ Alkalinity ( $\mu mol/L$ )	$\sigma_{Alkalinity}$	Alkalinity flux rate ( $10^6 mol/km^2/yr$ )	$\sigma_{Alkalinity flux rate}$	Latitude (°)	Longitude (°)	Holocene fraction (%)	Calculated Reactivity R
1	Massif Central	Inactive	8.770	0.65	406	20	916	46	0.372	0.026	45.7777	2.969600	0	0.0096
2	South Africa	Inactive	12.770	1.80	244	55	1728	1078	0.420	0.130	25.2827	29.63632	4	0.00
3	Karelia	Inactive	2.000	1.00	285	20	460	41	0.131	0.007	65.0000	31.00000	0	0.00
4	Coastal Deccan	Inactive	25.410	0.50	1690	150	657	17	1.110	0.103	16.9393	73.54510	0	0.00
5	Interior Deccan	Inactive	25.440	0.50	401	48	2839	170	1.138	0.152	21.0000	74.00000	0	0.00
6	Siberia Traps	Inactive	8.550	0.65	254	25	501	89	0.127	0.019	65.0000	100.0000	0	0.00
7	E'Mei	Inactive	6.220	1.00	1350	75	238	23	0.321	0.036	27.4544	103.3332	62	55
8	Lei-Qiong	Inactive	24.000	1.00	797	100	1923	165	1.532	0.233	20.4646	110.1818	00	0.00
9	Nanjing	Inactive	15.220	1.00	330	48	1595	63	0.526	0.079	32.7474	118.3939	00	0.00
10	Xiaoxinganling	Inactive	1.000	1.00	243	50	1065	132	0.259	0.062	49.0909	128.4717	42	04
11	Tumen River	Inactive	4.000	2.00	273	50	763	50	0.208	0.041	42.5050	128.5050	00	0.00
12	Mudan River	Inactive	3.220	2.00	209	46	977	87	0.204	0.048	43.7575	128.7272	00	0.00

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13	SE Australia	Inactive	13.00 0	0.10	74	14	5956	657	0.441	0.097	38.19 18 83	142.86 85 87	0.00
14	Tasmania	Inactive	10.41 0	0.25	221	30	1704	437	0.377	0.109	42.19 19 00	146.76 76 00	0.00
15	North Island, NZ	Inactive	13.00 0	2.00	920	161	478	131	0.439	0.143	38.00 00	176.00 00	0.00
16	Kauai, Hawaii	Inactive	21.65 8	0.65	1747	539	588	251	1.026	0.303	22.00 00	159.50 50 00	0.00
17	Columbia Plateau	Inactive	7.44 0	2.00	204	191	927	215	0.189	0.060	44.00 00	118.50 50 00	0.00
18	NE America	Inactive	0.77 0	2.30	507	129	465	316	0.235	0.075	47.05 04 83	75.32 32 9	0.00
19	Madeira Island	Inactive	13.55 0	1.50	1065	100	580	43	0.618	0.074	32.80 80 00	17.00 00 00	0.00
20	Easter Island	Active	20.66 0	1.00	580	100	1306	132	0.757	0.151	27.42 12 00	109.37 37 00	3.36
21	Mt. Cameroon	Active	14.00 0	2.00	2120	500	2368	110	5.020	1.207	4.00 00 00	9.00 00 00	96.57
22	Mt. Etna	Active	14.99 0	0.20	640	80	9286	539	5.943	0.819	37.75 75 14	15.00 14 9974	79.56
23	Virunga	Active	20.88 0	1.30	1709	380	2646	57	4.522	2.110	-1.50 00	29.50 50 00	48.0 247.9 5
24	La Réunion	Active	17.00 0	2.00	1712	652	1243	290	2.127	0.031	21.12 12 00	55.54 54 00	28.1 327.7 3
25	Wudalianchi Lake	Active	1.00 0	1.00	243	50	1919	190	0.466	0.106	48.70 70 10	126.14 14 11	14.82

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26	Japan	Active	11.0 10.99	1.12	1236	64	584	73	0.722	0.107	35.9292 34	135.3635 60	8.707
27	Kamchatka	Active	3.550 2.00		520	50	854	100	0.444	0.067	55.0000 00	159.0000 00	2.28
28	Taranaki	Active	10.00 0	3.00	1296	223	667	34	0.864	0.155	39.3030 00	174.0000 00	8.00
29	Big Island, Hawaii	Active	15.44 4	2.00	935	269	951	424	0.889	0.303	19.5050 00	155.5050 00	13.7 314.0 8
30	High Cascades	Active	6.881 0.47		382	172	776	175	0.296	0.108	45.1919 24	121.6868 44	0.202
31	Iceland São Miguel	Active	16.0 70	0.879 0.65	173423 34	37.7 7136	25.5 0498	749 35	0.864	0.109	65.0000	-18.0000	13.19
32	Sao Miguel Iceland	Active	0.71 6.00	1.00 1734	49887 9	65.0 050	18.0 0	13. 192	4.14 2.047	0.211	37.7700	-25.5000	9.35
33	Tianchi Lake	Active	7.330 2.00		291*	100	2445	300	0.711	0.260	42.0000 00	128.0505 00	65.34

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*\*The runoff for Tianchi Lake was recalculated after Fekete et al. (2002).*

9

10 **Supplemental Material B: New data for active volcanic fields as described in Table**  
11 **S1, in addition to (Li et al., 2016)**

12 In the following, all active volcanic provinces used for this study are described. Note, that  
13 for all inactive fields a detailed description of the input data can be found in Li et al. (2016),  
14 as well as the origin of the temperature, runoff and alkalinity/DIC concentration data for  
15 active and inactive volcanic fields. If watersheds are considered for the calculations of the  
16 Holocene area, they are based on the locations of the sampling points (see Li et al.  
17 (2016)).

18

19 **In the following, the new data for active volcanic fields are described (Table S1)**

20 For the calculation of the Holocene fraction area for each volcanic field we used several  
21 time spans. A polygon with an age description of Holocene (0-11.7 ka) was classified as  
22 a “Holocene”-area. If a polygon had an age description which laid in the time interval of 0  
23 to 2.58 Ma, but it was not clearly defined as of Holocene or Pleistocene age, we defined  
24 it as “Quaternary” and applied a theoretical Holocene fraction by the ratio of Holocene  
25 time span and Quaternary time span. All other polygons with age descriptions older than  
26 Holocene and not defined as “Quaternary” were defined as “Non-Holocene”.

27 Numbers behind a region indicate the number in the summary table S1.

28

29 **Easter Island (No.20)**

30 Easter Island is a volcanic island in the eastern Pacific. Its volcanic nature is related to  
 31 the Easter Island Hotspot. The geologic map used for this study was digitized after a map  
 32 of Gioncada et al. (2010) and references therein. The island is composed mostly of  
 33 hawaiites, mugearites and olivine basalts and it comprises volcanic ages from 3 Ma to  
 34 recent. The different age classifications and lithologies are listed in the table below. Due  
 35 to the precise age information the rocks ~~could be~~ directly classified into Holocene  
 36 rocks and non-Holocene rocks. The total basaltic area of the island is 162.2827 km<sup>2</sup>, the  
 37 Holocene area 5.45 km<sup>2</sup> and the non-Holocene area 156.82 km<sup>2</sup>, resulting in a Holocene  
 38 area fraction of 3.36%.

39

*Table S2: Classification of the map data of Easter Island. The first three columns provide the original map data, whereas the column "System/Series" shows our interpretation of the map data.*

Name	Description	Age	System/Series
TE1	Terevaka, hawaiites and olivine basalts	<del>0.3-1.9-0.3</del> Ma	non-Holocene
TE2	Terevaka, hawaiites and olivine basalts	<del>0.3-1.9-0.3</del> Ma	non-Holocene
TE3	Terevaka, hawaiites and olivine basalts	<del>0.3-1.9-0.3</del> Ma	non-Holocene
HH2	Anakena, Hiva-Hiva, hawaiites and olivine basalts	0- <del>2000</del> <u>2,000</u> a ago	Holocene
RA1	Rano Aroi, hawaiites, olivine basalts and mugearites	0.2 Ma	non-Holocene
RA5	Rano Aroi, hawaiites, olivine basalts and mugearites	0.2 Ma	non-Holocene
RA4	Rano Aroi, hawaiites, olivine basalts and mugearites	0.2 Ma	non-Holocene
RA2	Rano Aroi, hawaiites, olivine basalts and mugearites	0.2 Ma	non-Holocene
RA8	Rano Aroi, hawaiites, olivine basalts and mugearites	0.2 Ma	non-Holocene
RA7	Rano Aroi, hawaiites, olivine basalts and mugearites	0.2 Ma	non-Holocene
RA6	Rano Aroi, hawaiites, olivine basalts and mugearites	0.2 Ma	non-Holocene
RA3	Rano Aroi, hawaiites, olivine basalts and mugearites	0.2 Ma	non-Holocene

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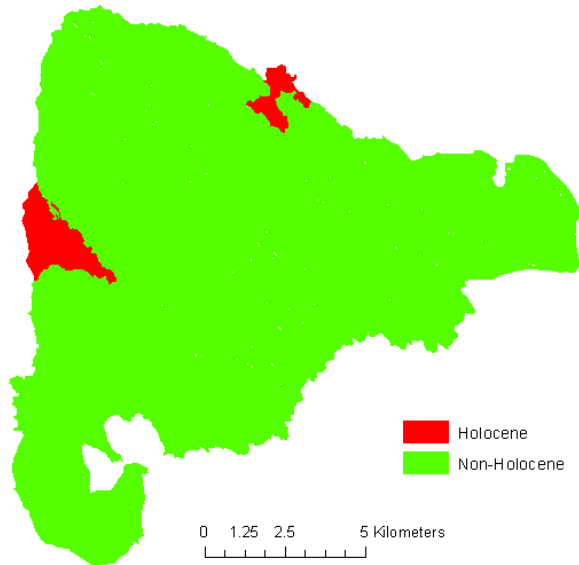
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HH1	Anakena, Hiva-Hiva, hawaiites and olivine basalts	0- <del>2000</del> <u>2,000</u> a ago	Holocene
PO2	Polke, alkali basalts, hawaiites to mugearites	<del>3</del> -0.61- <del>3</del> Ma	non-Holocene
PO1	Polke, alkali basalts, hawaiites to mugearites	<del>3</del> -0.61- <del>3</del> Ma	non-Holocene
PO5	Polke, alkali basalts, hawaiites to mugearites	<del>3</del> -0.61- <del>3</del> Ma	non-Holocene
PO4	Polke, alkali basalts, hawaiites to mugearites	<del>3</del> -0.61- <del>3</del> Ma	non-Holocene
TR	Maunga Orito, rhyolite; Maunga Parehe, trachyte	not known	not considered
PO3	Polke, alkali basalts, hawaiites to mugearites	<del>3</del> -0.61- <del>3</del> Ma	non-Holocene
RK1	Rano Kau, alkali basalts, hawaiites to benmoreites	<u>0.2</u> -2.5- <del>0.2</del> Ma	non-Holocene
RK2	Rano Kau, alkali basalts, hawaiites to benmoreites	<u>0.2</u> -2.5- <del>0.2</del> Ma	non-Holocene
TA4	Tangaroa, hawaiites, olivine basalts and mugearites	0.2 Ma	non-Holocene
TA1	Tangaroa, hawaiites, olivine basalts and mugearites	0.2 Ma	non-Holocene
TA2	Tangaroa, hawaiites, olivine basalts and mugearites	0.2 Ma	non-Holocene
TA6	Tangaroa, hawaiites, olivine basalts and mugearites	0.2 Ma	non-Holocene
TA3	Tangaroa, hawaiites, olivine basalts and mugearites	0.2 Ma	non-Holocene
TA5	Tangaroa, hawaiites, olivine basalts and mugearites	0.2 Ma	non-Holocene
TA7	Tangaroa, hawaiites, olivine basalts and mugearites	0.2 Ma	non-Holocene

40

41



42

Figure S1: Map of Easter Island showing the Holocene and non-Holocene areas.

Table S3: Calculated areas for Easter Island. *The Holocene fraction is derived by the ratio of Holocene area/Total area.*

Area Holocene (km <sup>2</sup> )	Area Non-Holocene (km <sup>2</sup> )	Area Quaternary (km <sup>2</sup> )	Total Area (km <sup>2</sup> )	Holocene (%)
5.45	156.8382	0	162.2827	3.36

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45 **Mount Cameroon (No.21)**

46 Mount Cameroon is part of a volcanic chain at the coast of West Africa. The geological  
 47 map was digitized from Le Maréchal (1975). ~~The classification of the individual~~  
 48 ~~theologies~~ The location of the sampling points were derived from Benedetti et al. (2003).  
 49 The age classification (see table below) results in a Holocene fraction of 96.57%.

Table S4: Classification of the ~~theologies~~ surface ages of Mt. Cameroon. Note that the first three columns display the original map data, the column "System/Series" provides the authors interpretation.

Name	Description	Age	System/Series
beta1	Séries inférieures: basaltes parfois andésitiques sous forme de coulées et de dykes	Oligocene- <u>Eocene</u>	non-Holocene
beta3	Séries supérieures: basaltes parfois andésitiques sous forme de coulées et cinérites	quaternaire récente	Holocene

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50  
 51 The interpretation of the ages of Mount Cameroon was difficult, since the ages are not  
 52 well determined (Ateba et al., 2009). Le Maréchal (1976) describes the ages of the  
 53 basalts, as well as an unpublished map in Ateba et al. (2009) that shows "young basalts"  
 54 in the center of Mount Cameroon so that we assumed an age of "recent quaternary" for  
 55 "beta3".

Table S5: Area calculation of Mt. ~~Cameroon~~ Cameroon. The Holocene fraction is derived by the ratio of Holocene area/Total area.

Area Holocene (km <sup>2</sup> )	Area non-Holocene (km <sup>2</sup> )	Area Quaternary (km <sup>2</sup> )	Total Area (km <sup>2</sup> )	Holocene (%)
<del>444</del> 21,112.63	39.48	0	<del>445</del> 21,152.11	96.57

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58

*Figure S2: Map of Mount Cameroon showing the Holocene and non-Holocene area.*

59

60 **Mount Etna (No.22)**

61 ~~Mount Etna is located at a subduction zone in Italy. Its geological map was digitized from~~  
 62 ~~Branca et al. (2011). The detailed description of the lithologies can be seen below and~~  
 63 ~~provides a classification into Holocene, non-Holocene and Quaternary.~~

64 **Mount Etna (No.22)**

65 Mount Etna is located at a subduction zone in Italy. Its geological map was digitized from  
 66 Branca et al. (2011a) and is described in Branca et al. (2011b). The detailed description  
 67 of the lithologies is shown below and allows for a classification into Holocene, non-  
 68 Holocene and Quaternary.

69 The Quaternary rocks are of an age of ~~15ka~~3.9 ka to ~~3.9ka~~15 ka, so that they have a time  
 70 span of ~~11.4ka~~1 ka. ~~7.8 ka are within the Holocene period (3.9 ka to 11.7~~  
 71 ~~ka) considering the Holocene time period going from 0 to 11.7 ka the amount . The fraction~~  
 72 ~~of Holocene coverage can be was than~~ calculated by:

73 ~~Holocene fraction = (Area Holocene + (Area Quaternary x (7.8ka / 11.1ka))) /~~  
 74 ~~Total Area~~

75 
$$Holocene\ fraction = \frac{Area\ Holocene + \left( Area\ Quaternary \times \left( \frac{7.8}{11.1} \right) \right)}{Total\ Area}$$

77 resulting in a Holocene fraction area of 79.56%.

Table S6: Classification of Etna basaltic rocks showing the original map data (first four columns) and our interpretation of the data ("System/Series").

Symbol	Name	Description	Age	System/Series
26u	lava flows, cinder cones and bastions, and fall deposits	basaltic to benmoreitic	<del>4ka</del> - 122 b.C. - <del>4 ka</del>	Holocene
27-1	Castings, cinder cones and bastions, and fall deposits	basaltic to mugaritica	<del>1669</del> - 122 b.C. - <del>1669</del>	Holocene
27-2	Castings, cinder cones and bastions, and fall deposits	basaltic to mugaritica	<del>1669</del> - <del>1971</del>	Holocene
27-3	Lava flows, cinder cones and bastions, and fall deposits	basaltic to mugaritica	<del>1971</del> - current - <del>1971</del>	Holocene
CB26u	Cono e bastione di scorie			Holocene
CB27-1	Cono e bastione di scorie			Holocene
CB27-2	Cono e bastione di scorie			Holocene
CB27-3	Cono e bastione di scorie			Holocene

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10a	: intercalated flows in a powerful pyroclastic flow deposit	mugearitica	nn	non-Holocene
14	Succession of alternating flows in pyroclastic deposits	hawaiiitica to mugearitica	nn	non-Holocene
15	flows intercalated with clastic deposits	mugearitica to benmoreitic	nn	non-Holocene
17	Casting interbedded with clastic deposits	benmoreite	nn	non-Holocene
18	flows	mugearitica-benmoreitic	nn	non-Holocene
18a	bodies subvolcanici materials come from lavas	mugearitica-benmoreitic	nn	non-Holocene
1a	bodies subvolcanici		nn	non-Holocene
21b	porfirichi flows	hawaiiitica to mugearitica	nn	non-Holocene
22b	casting succession	hawaiiitica	nn	non-Holocene
24	of breccia autoclastic often altered to hydrothermalism, associated with lava flows	benmoreitic	nn	non-Holocene
26b	deposit of debris avalanche monogenic, formed by lava mugearitica blocks	nn	nn	Quaternary
3b	massive lava flows	basaltic	nn	non-Holocene
1	underwater transitional composition of volcanics in tholeiitica consist of pillow lavas	tholeiitica	<del>496.1 - 542.2- 496.1ka ka</del>	non-Holocene
10b	flows intercalated with breccias autoclastic and deposits epiclastici	hawaiiitica to mugearitica	101.9ka	non-Holocene
11	Brecce autoclastic and deposits epiclastici	benmoreitic	<del>99.1 - 107.2- 99.1ka ka</del>	non-Holocene
12	deposits associated with pyroclastic flows	mainly mugearitica	<del>99.9 - 101.8- 99.9ka ka</del>	non-Holocene
13	castings and slag deposits	hawaiiitica	93.0ka0 ka	non-Holocene
14a	subvolcanici bodies formed by lava massive	mugearitica	<del>85.3ka3 ka</del>	non-Holocene
16	: Casting intercalated with thin epiclastici deposits	mugearitica to benmoreitic	85.6ka6 ka	non-Holocene
19	Thin flows to the base, followed by a thick succession pyroclastic	mugearitica	<del>70.2 - 79.6- 70.2ka ka</del>	non-Holocene
2	lava flows	tholeiitica	332.4ka4 ka	non-Holocene

20	Fall and pyroclastic flow deposits	hawaiiitica to benmoreitic	<del>41.3 - 56.6</del> 41.3ka ka	non-Holocene
21a	massive flows and autoclastic interbedded with breccia deposits	hawaiiitica to mugaritica	29.1- <del>32.5ka</del> 5 ka	non-Holocene
22	castings, scoria cones and fall deposits	hawaiiitica to benmoreitic	28.7- <del>42.1ka</del> 1 ka	non-Holocene
<del>22</del>	<del>eastings, scoria cones and fall deposits</del>	<del>hawaiiitica to benmoreitic</del>	<del>28.7- 42.1ka</del>	<del>non-Holocene</del>
<del>22</del>	<del>eastings, scoria cones and fall deposits</del>	<del>hawaiiitica to benmoreitic</del>	<del>28.7- 42.1ka</del>	<del>non-Holocene</del>
<del>22</del>	<del>castings, scoria cones and fall deposits</del>	<del>hawaiiitica to benmoreitic</del>	<del>28.7- 42.1ka</del>	<del>non-Holocene</del>
<del>22</del>	<del>castings, scoria cones and fall deposits</del>	<del>hawaiiitica to benmoreitic</del>	<del>28.7- 42.1ka</del>	<del>non-Holocene</del>
<del>22</del>	<del>castings, scoria cones and fall deposits</del>	<del>hawaiiitica to benmoreitic</del>	<del>28.7- 42.1ka</del>	<del>non-Holocene</del>
<del>22</del>	<del>castings, scoria cones and fall deposits</del>	<del>hawaiiitica to benmoreitic</del>	<del>28.7- 42.1ka</del>	<del>non-Holocene</del>
25b	reomorfiche flows	benmoreitic	15.0 - 15.4- <del>15.0ka</del> ka	non-Holocene
2a	subvolcanico body structure		320.0ka <del>0 ka</del>	non-Holocene
3a	strongly altered lava flows	basaltic	180.2ka <del>2 ka</del>	non-Holocene
4a	massive lava flows	basaltic to mugaritica	129.9 - 154.9- <del>129.9ka</del> ka	non-Holocene
4b	thin lava flows	basaltic to mugaritica	134.2ka <del>2 ka</del>	non-Holocene
6	Lave cataclasate	basaltic to mugaritica	128.7ka <del>7 ka</del>	non-Holocene
7	Succession Lava	mugaritica	126.4ka <del>4 ka</del>	non-Holocene
8	Succession lava with thin interbedded deposits epiclastici	hawaiiitica to mugaritica	111.9 - 121.2- <del>111.9ka</del> ka	non-Holocene
9	lava flows interbedded with massive deposits of local epiclastici	hawaiiitica to benmoreitic	105.8ka <del>8 ka</del>	non-Holocene
CB2	Cono e bastione di scorie			non-Holocene
CB22	Cono e bastione di scorie			non-Holocene
CB3a	Cono e bastione di scorie			non-Holocene
CB7	Cono e bastione di scorie			non-Holocene

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26I	lava flows, cinder cones and bastions, and fall deposits	basaltic to benmoreitic	<del>4 - 15ka</del> 4 ka	Quaternary
CB26I	Cono e bastione di scorie			Quaternary

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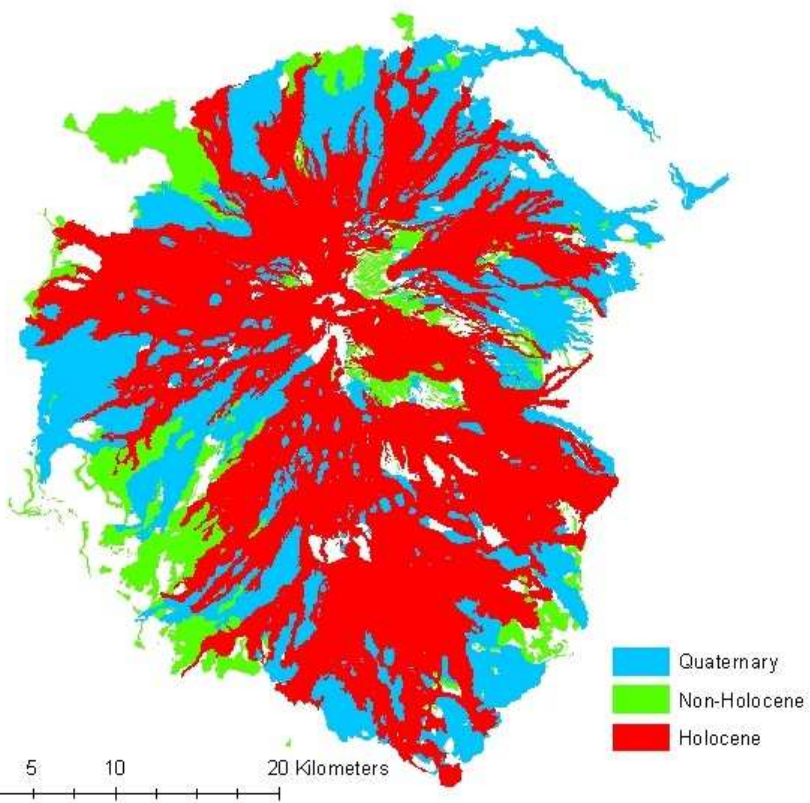
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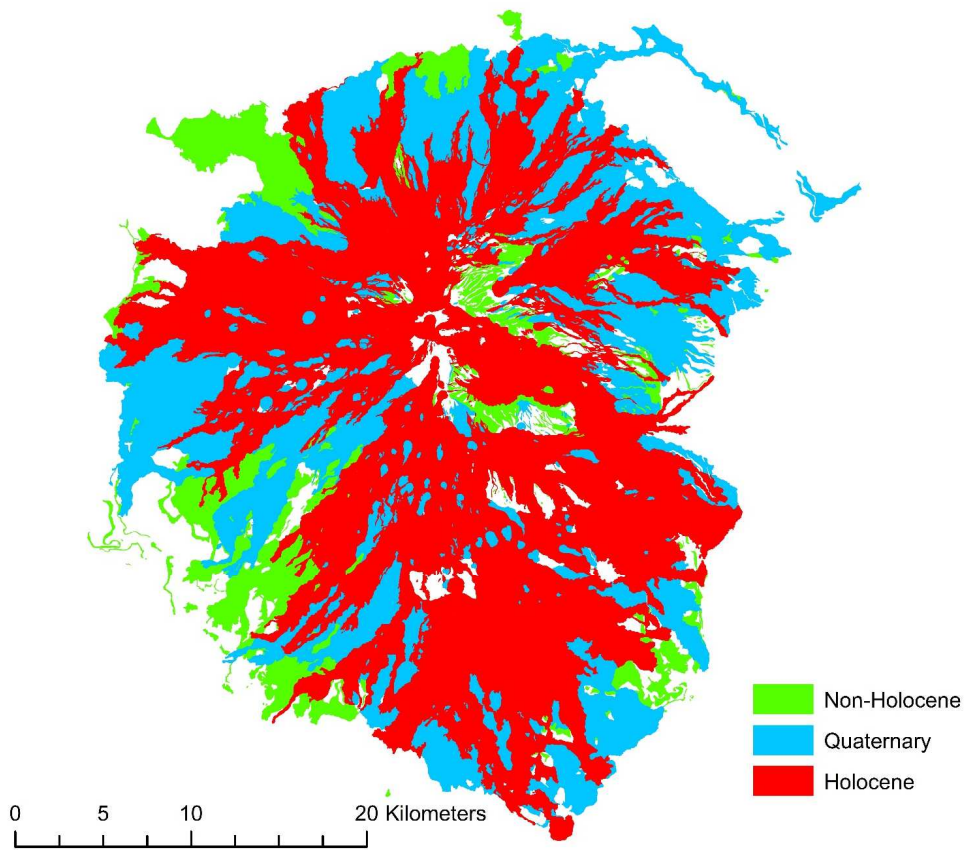
Table S7: Table showing the calculated areas for Mount Etna. *Note that the Holocene fraction is calculated by the above mentioned equation considering Holocene area, Quaternary area and total area.*

Area Holocene (km <sup>2</sup> )	Area non-Holocene (km <sup>2</sup> )	Area Quaternary (km <sup>2</sup> )	Total Area (km <sup>2</sup> )	Holocene (%)
636.16	123.90	338.39	<del>1098.46</del> 1,098.45	79.56

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83  
*Figure S3: Map of Etna volcano showing the age distribution of basaltic rocks.*

84 **Virunga (No.23)**

85 The Virunga Volcanic province is located in eastern central Africa and is related to the  
 86 East African Rift system. The geologic map of this area was compiled ~~out of~~ from four  
 87 different maps ~~from Smets et al. (2010)~~ published by Antun et al. (1971), Balagizi et al.  
 88 (2015) De Mulder et al. (1986), ~~Petricc et al. (1971)~~ Smets et al. (2010), and ~~De~~  
 89 ~~Mulder~~ Balagizi et al. (1986, 2015).

90 The western part of the province is classified as purely Holocene, whereas the volcanic  
 91 rocks become older to the east. Considering the Quaternary age period ranging from 0 to  
 92 2.58Ma, the Holocene percentage of the Quaternary area is calculated by the ratio of  
 93 11700a/2580000a. The total theoretical Holocene areal percentage is 48.02% using:

94 
$$\text{Holocene fraction} = \frac{\text{Area Holocene} + (\text{Area Quaternary} \times (11700a / 2580000a))}{\text{Total Area}}$$

96 We assume that the volcanic areas of Nyiragongo and Nyamuragira are of Holocene age,  
 97 since they are classified as Africa's most active volcanoes, and erupted frequently in the  
 98 past (Smets et al., 2010; Balagizi et al., 2015). The remaining volcanoes are assumed to  
 99 be of quaternary age (not older than Pleistocene (Antun et al., 1971). For the northeastern  
 100 part of the map the surface age could not be determined clearly, and was therefore  
 101 defined as "not known", and included in the calculations as "non-Holocene".

102 Considering that the Quaternary age period ranges from 0 to 2.58 Ma, the Holocene  
 103 percentage of the Quaternary area is calculated by the ratio of 11,700 a / 2,580,000 a.  
 104 The total theoretical Holocene areal percentage is 47.95% using:

105 
$$\text{Holocene fraction} = \frac{\text{Area Holocene} + \left( \text{Area Quaternary} \times \left( \frac{11,700}{2,580,000} \right) \right)}{\text{Total area}}$$

106

Table S8: Classification of basaltic rocks in the Virunga province showing the individual volcanoes and our interpretation of their age.

<b>Id</b>	<b>Volcano</b>	<b>System/Series</b>
0	Nyiragongo	Holocene
1	Nyamuragira	Holocene
2	Muhabura	<del>Pleistocene</del> Quaternary
3	Gahinga	Quaternary <del>Pleistocene</del>
4	<del>Muhabura-Gahinga</del> Synabaye	Quaternary <del>Pleistocene</del>
5	<del>Bisoke-Sabinyo</del>	Quaternary <del>Pleistocene</del>
6	<del>Bisoke-Sabinyo-Gahinga</del>	<del>Pleistocene</del> Quaternary
7	Bisoke-Sabinyo	Quaternary
8	Karisimbi	Quaternary

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9	Remaining areas	<del>Quaternary</del> Not known
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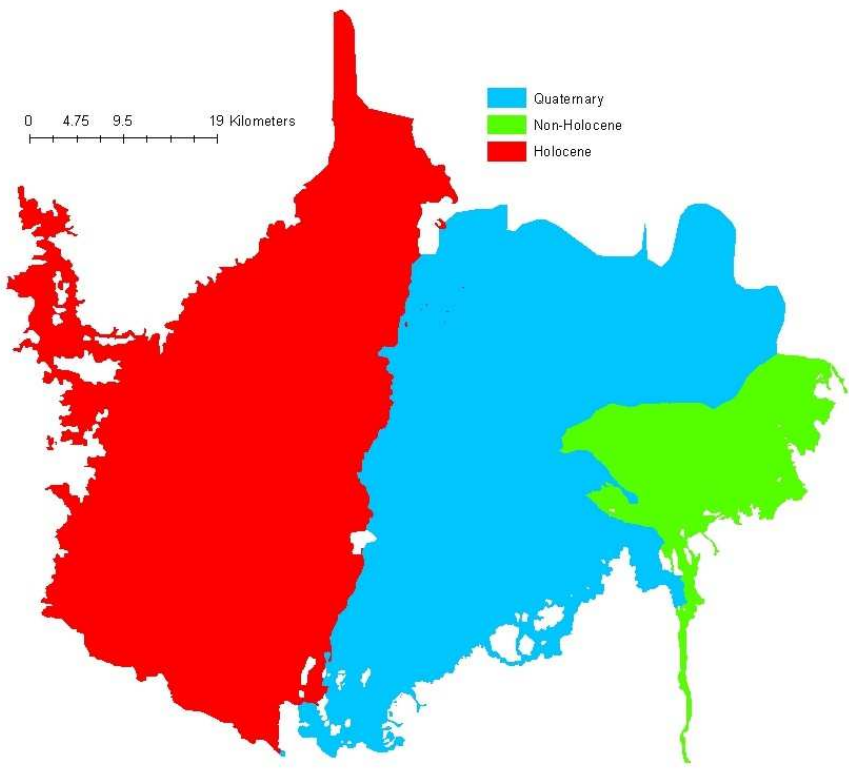
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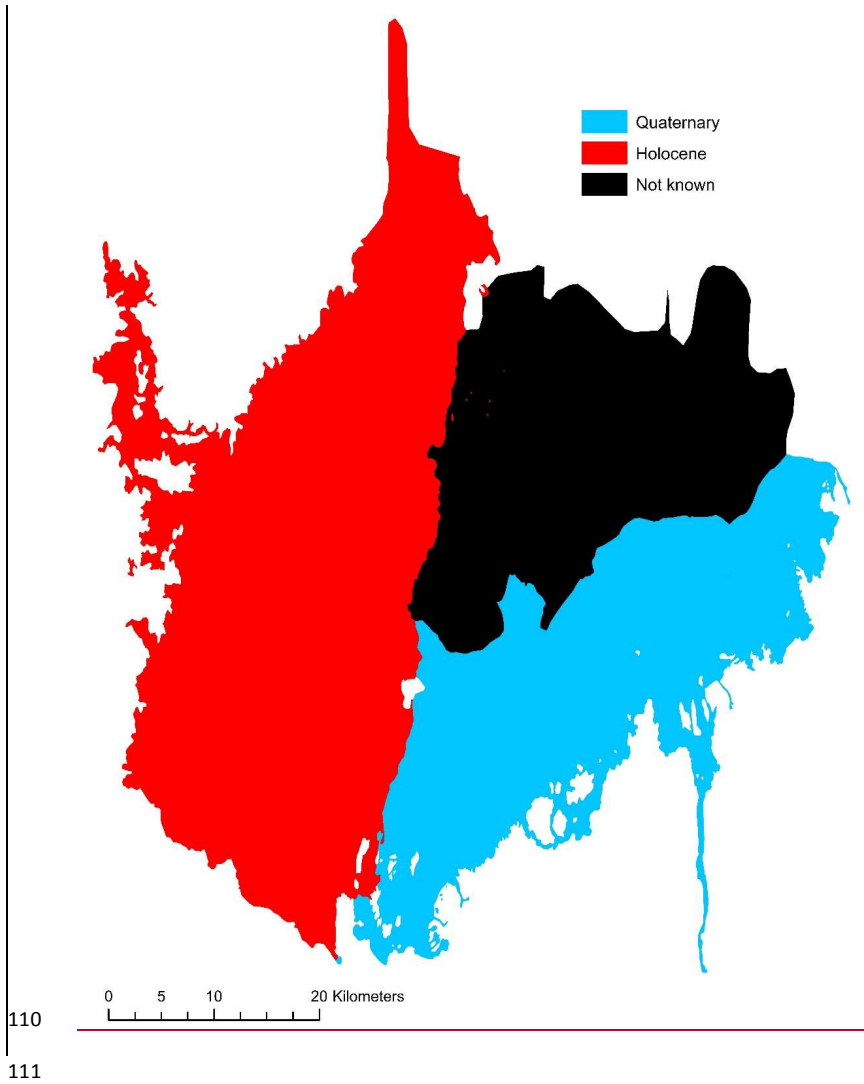
Table S9: Calculated areas for Virunga province. *The Holocene fraction is derived by applying the above mentioned equation considering the Holocene area, Quaternary area and total area.*

Area Holocene (km <sup>2</sup> )	Area Non-Holocene (km <sup>2</sup> )	Area Quaternary (km <sup>2</sup> )	Total Area (km <sup>2</sup> )	Holocene (%)
15181,518.62	320.69791.26	1335.72865.16	3175.033,175.04	48.0247.95

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110  
111  
*Figure S4: Map of the Virunga province showing the different areas of basalt classified after age.*

112

113 **La Réunion (No.24)**

114 The volcanic island La Réunion belongs to a Hot Spot in the Indian Ocean and is  
 115 comprised of Holocene, ~~non-Holocene and Quaternary volcanic rocks. The geological~~  
 116 ~~map was digitized from Nehlig et al. (2006). The rocks can be divided into Holocene, non-~~  
 117 ~~Holocene and two different types of Quaternary rocks (Qu1, Qu2); Quaternary and older~~  
 118 volcanic rocks. The geological map was digitized from Nehlig et al. (2006). The watershed  
 119 area considered for this study is derived from Louvat and Allègre (1997). The rocks are  
 120 divided into Holocene, non-Holocene and two different types of Quaternary rocks (Qu1,  
 121 Qu2).

122 ~~Qu1: <340ka → is described as being younger than 340 ka which results in a theoretical~~  
 123 ~~Holocene fraction = of 11.7ka/340ka 7 ka / 340 ka,~~

124 Qu2: 65ka – 5ka → Holocene fraction = 6.7ka/60ka

125 The age of Qu2 ranges from 5 ka to 65 ka, providing a time span of 60 ka. 6.7 ka lay  
 126 within the Holocene time span (11.7 ka – 5 ka), so that the Holocene fraction can be  
 127 calculated by the ratio of 6.7 ka / 60 ka.

128 Thus, the total Holocene fraction ~~can be derived~~ is calculated by:

129 Holocene fraction = (Area Holocene + (Area Qu1 x (11.7ka / 340ka)) + (Area Qu2  
 130 x (6.7ka / 60ka))) / Total Area

131 
$$\text{Holocene fraction} = \frac{\left(\text{Area Holocene} + \left(\text{Area Qu1} \times \left(\frac{11.7}{340}\right)\right) + \left(\text{Area Qu2} \times \left(\frac{6.7}{60}\right)\right)\right)}{\text{Total area}}$$

132 providing a value of 28.13%.

133 *Table S10: Classification of the basaltic rocks of La Réunion with the original data (first three columns) and our interpretation ("System/Series"). Note that "nn" represents that the age of the lithology is not known.*

ID	Definition	Type	System/Series
beta 4	Coulées basaltiques	Massif du Piton de La Fournaise - Série du <del>bouelle#bouclier</del> ancien (450000450,000 à 450000150,000 ans)	non-Holocene
tfp	Pitons et projections	Massif du Piton de La Fournaise	nn
beta 7	Coulées basaltiques	Massif du Piton de La Fournaise - Série de la Plaine des Cafres ( <del>6500065,000</del> à <del>50005,000</del> ans)	Quaternary2

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beta 6	Coulées basaltiques	Massif du Piton de La Fournaise - Série Plaine des Sables ( <del>65000</del> 65.000 à <del>5000</del> 5.000 ans)	Quaternary2
beta 8	Coulées basaltiques	Massif du Piton de La Fournaise - Série volcanique subactuelle (< <del>5000</del> 5.000 ans)	Holocene
beta 5	Coulées basaltiques	Massif du Piton de La Fournaise - Série des Remparts ( <del>150000</del> 150.000 à <del>65000</del> 65.000 ans)	non-Holocene
beta 3	Coulées différenciées	Massif du Piton de La Fournaise - Série alcaline anté-Fournaise ( <del>530000</del> 530.000 à <del>450000</del> 450.000 ans)	<del>non-Holocene</del> Not considered
beta 8e	Coulées basaltiques dans l'Enclos	Massif du Piton de La Fournaise - Série volcanique subactuelle (< <del>5000</del> 5.000 ans)	Holocene
beta 1	Coulées basaltiques à olivine	Massif du Piton des Neiges - Série des océanites (> <del>340000</del> 340.000 ans)	non-Holocene
beta 2	Coulées (basalte, hawaïtes, mugéarites)	Massif du Piton des Neiges - Série différenciée (< <del>340000</del> 340.000 ans)	Quaternary1
Br	Brèches d'avalanches de débris de Saint Gilles	Massif du Piton des Neiges - Série différenciée (< <del>340000</del> 340.000 ans)	Quaternary1
pc	Coulées ignimbritiques	Massif du Piton des Neiges - Série différenciée (< <del>340000</del> 340.000 ans)	Quaternary1
Tau	Coulées trachytiques du plateau de Belouve	Massif du Piton des Neiges - Série différenciée (< <del>340000</del> 340.000 ans)	Quaternary1 not considered

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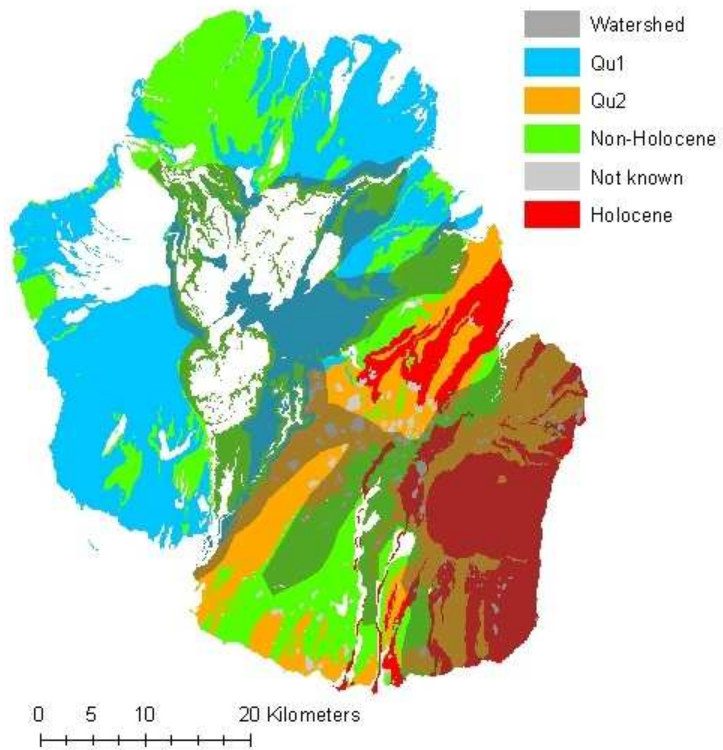
Table S11: Summary of calculated areas of La Réunion. *Note that "tfp" is considered as of non-Holocene age. The Holocene fraction is calculated by the above mentioned equation considering Holocene area, Qu1 area, Qu2 area and the total area.*

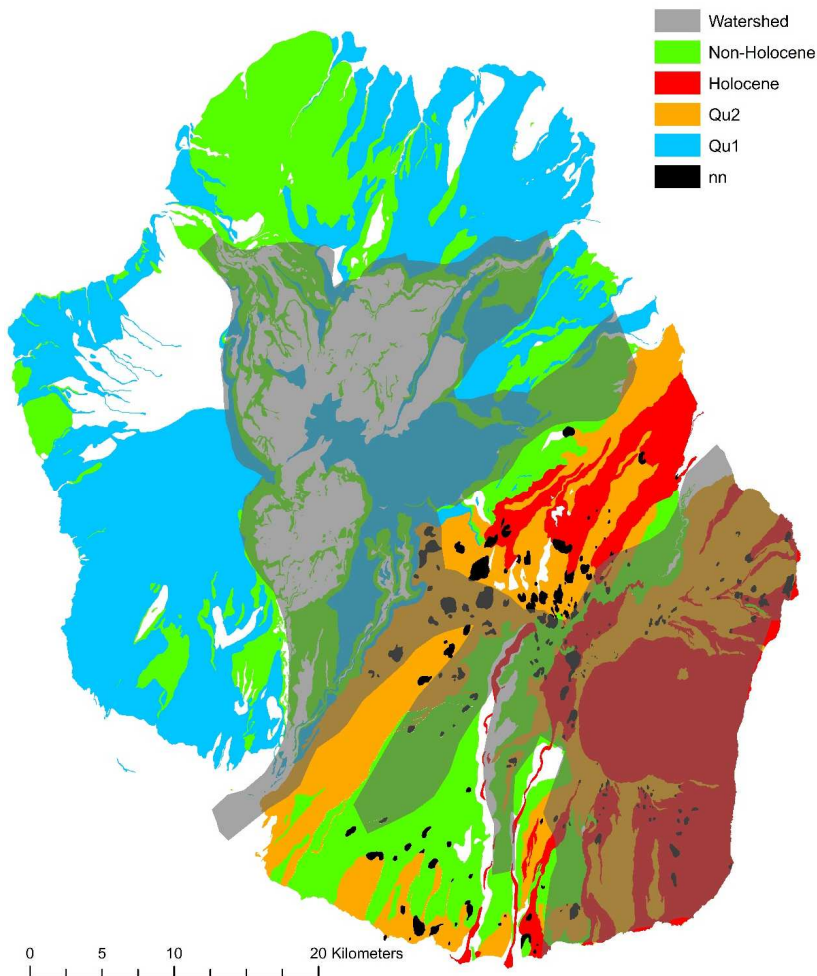
Area Holocene (km <sup>2</sup> )	Area non-Holocene (km <sup>2</sup> )	Area Qu1 Quaternary (km <sup>2</sup> )	Area Qu2 (km <sup>2</sup> )	Total Area (km <sup>2</sup> )	Holocene (%)
208.11	<del>270.98</del> 285.2	<del>359.60</del> 157.71	<del>838.68</del> 199.3	<del>28.13</del> 850.4	<del>27.73</del>

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Figure S5: Map of La Réunion showing the basaltic ages and additionally the watershed of the water samples. Only the watershed area was considered for this study.

139

140 **Wudalianchi Lake (No.25)**

141 The basaltic rocks of Wudalianchi Lake are located in NE China and are of intraplate  
142 volcanic origin. The Geomap of Wudalianchi region was digitized after a map of the  
143 Ministry of Land and Resources of PRC, 2003 (unpublished work). The basalts ~~can be~~  
144 ~~separated into~~are of Holocene and non-Holocene ~~and thus provide a calculated~~age. The  
145 Holocene fraction ~~of~~is 14.82%.

146

Table S12: Classification of ~~basalt~~basalts of Wudalianchi Lake with the original map data (first three columns) and our interpretation shown in the column "System/Series".

Symbol	Litho	Age	System/Series
betaQ2w	basalt	middle pleistocene	non-Holocene
betaQ1j	basalt	lower pleistocene	non-Holocene
betaQ2b	basalt	middle pleistocene	non-Holocene
betaQ1g	basalt	tertiary	non-Holocene
betaQ42l	basalt	holocene	Holocene

147

Table S13: Calculated areas of Wudalianchi Lake. ~~The Holocene fraction is calculated~~ by Holocene area/total area.

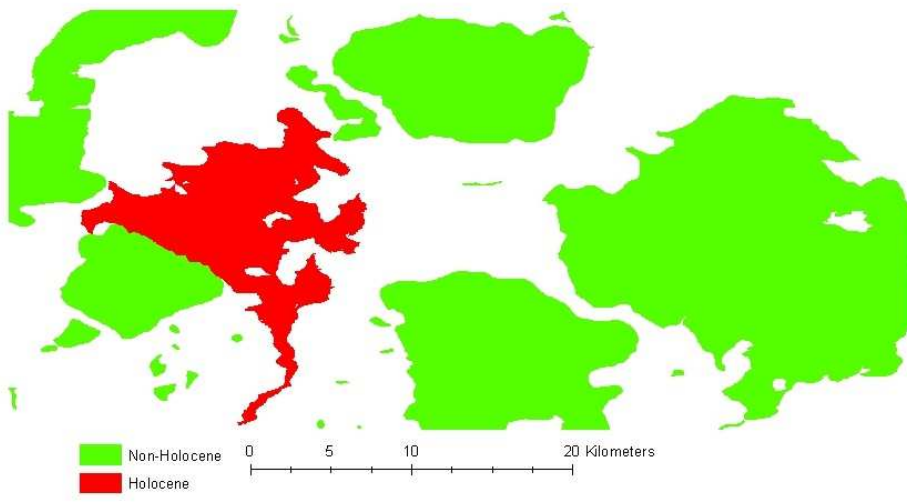
Area Holocene (km <sup>2</sup> )	Area non-Holocene (km <sup>2</sup> )	Area Quaternary (km <sup>2</sup> )	Total Area (km <sup>2</sup> )	Holocene (%)
69.68	400.35	0	470.0403	14.82

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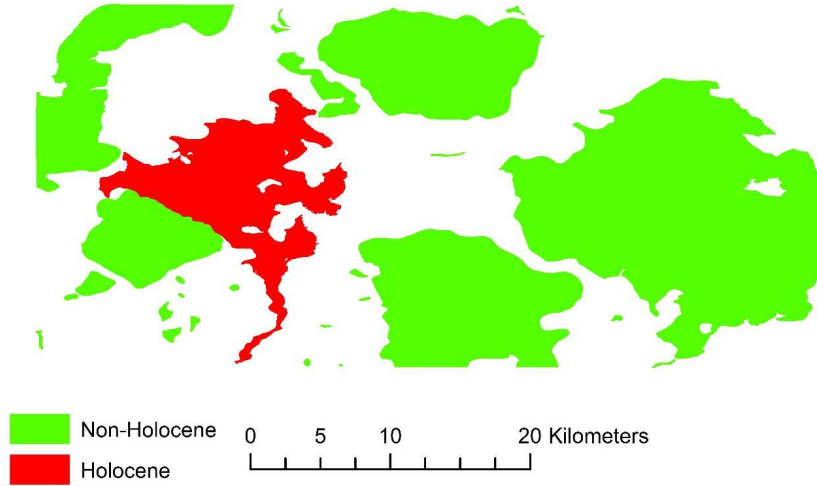


Figure S6: Map of Wudalianchi Lake showing the distribution of Holocene and non-Holocene basaltic rocks.

151

152

153 **Japan (No.26)**

154 The geological data was ~~derived of~~taken from the Global Lithological Map (GLiM) by  
 155 Hartmann and Moosdorf (2012) ~~but only the~~ Only watersheds of sample locations from  
 156 the GLORICH database (Hartmann et al., 2014) were used, ~~thus the~~ The basaltic rocks  
 157 ~~can be divided~~(attribute xx = 'vb' in the GLiM) are classified into Holocene, non-Holocene  
 158 and Quaternary rocks. The calculation procedure of the Holocene fraction of Quaternary  
 159 rocks is the same as for the Virunga Province, (No.23), and a final Holocene percentage  
 160 of 8.70% is calculated.

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Table S14: Classification of basaltic rocks for Japan after the GLiM (first three columns) and our interpretation "System/Series".

xx	Age_Min	Age_Max	System/Series
vb	Holocene	Holocene	Holocene
vb	Neogene	Neogene	non-Holocene
vb	Paleogene	Paleogene	non-Holocene
vb	Paleozoic	Proterozoic	non-Holocene
vb	Pleistocene	Pleistocene	non-Holocene
vb	Pliocene	Pliocene	non-Holocene
vb	Quaternary	Quaternary	Quaternary

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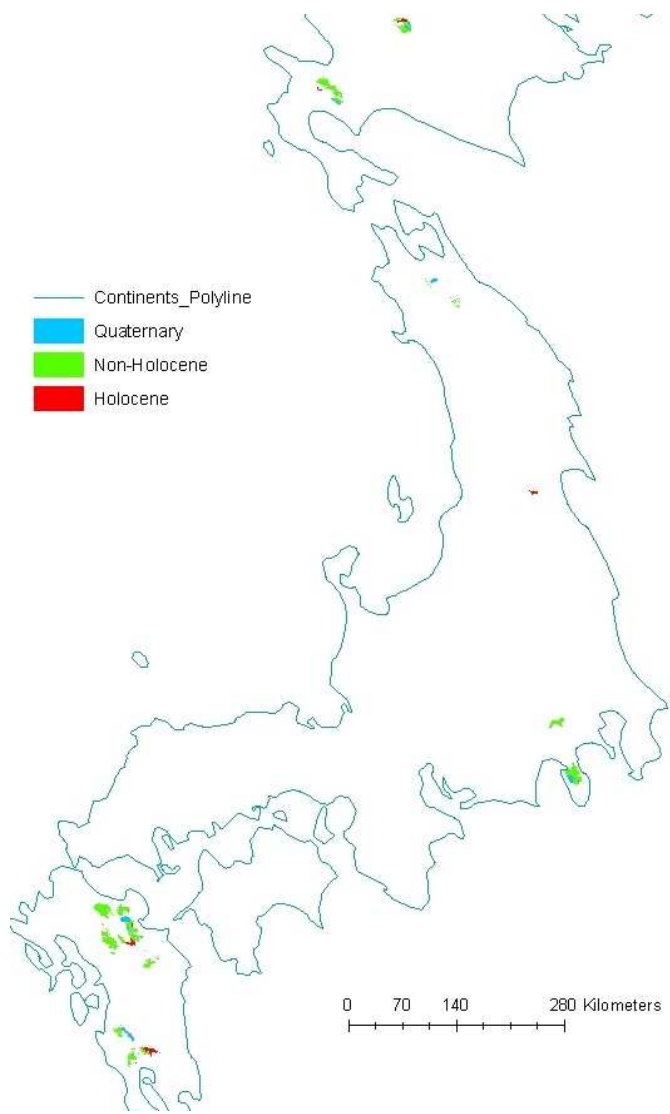
Table S15: Calculated areas for ~~Japan~~the basaltic watersheds in Japan. Note that the Holocene fraction is calculated considering Holocene area, Quaternary area and total area.

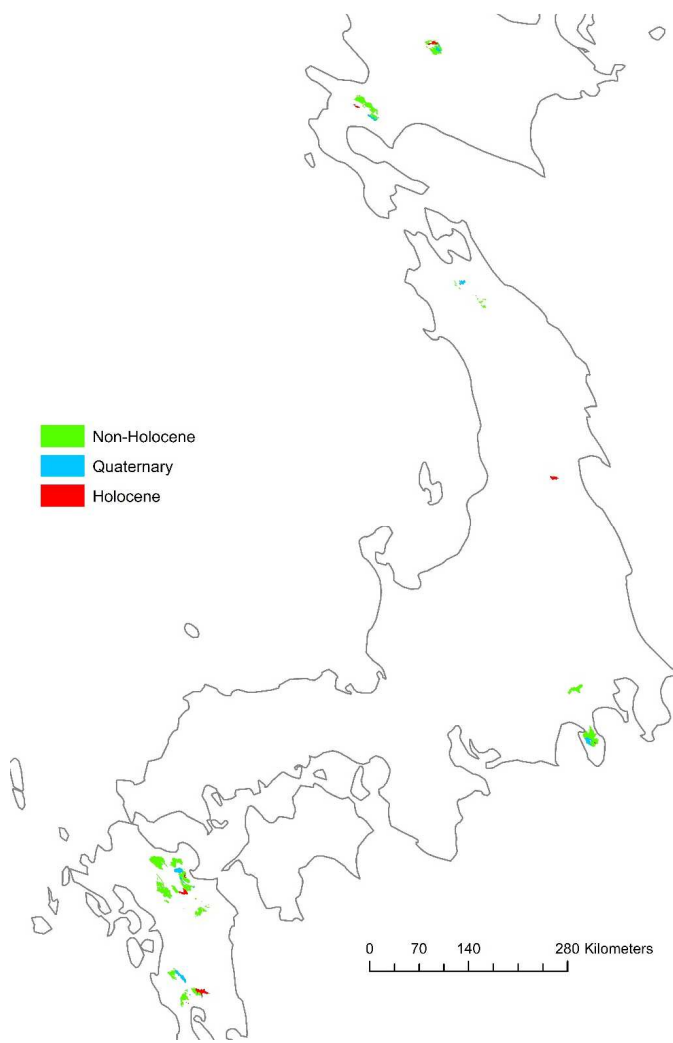
Area Holocene (km <sup>2</sup> )	Area non-Holocene (km <sup>2</sup> )	Area Quaternary (km <sup>2</sup> )	Total Area (km <sup>2</sup> )	Holocene (%)
243.30	<del>22162</del> 216.41	354.44	<del>28142</del> 814.15	8.70

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Figure S7: Map showing the watersheds and age classification of basaltic rocks for Japan.

168



169 **Kamchatka (No.27)**

170 The information of the basaltic rocks of the Kamchatka Peninsula is taken ~~out offrom~~ the  
 171 GLiM, attribute xx='vb' (Hartmann and Moosdorf, 2012) ~~and provideswith the~~ age  
 172 ~~information ofclasses~~ Holocene, non-Holocene and Quaternary, ~~where the.~~ The  
 173 Holocene fraction ~~is againwas~~ calculated as for the Virunga province, (No.23). The  
 174 watershed was calculated using sampling locations of Dessert et al. (2009). The  
 175 Holocene fraction of the watershed of the Kamchatka Peninsula is 2.28%.

176

Table S16: Classification of basaltic rocks of the Kamchatka Peninsula after the GLiM (first three columns) and our interpretation ("System/Series").

Rock_Description	Age_Min	Age_Max	System/Series
Basalte, Andesite und deren Tuffe	Lower Quaternary	Lower Quaternary	non-Holocene
Basalte, Andesite und deren Tuffe	Middle Quaternary	Middle Quaternary	non-Holocene
volcanogenic formations, basic composition	Pliocene	Pliocene	non-Holocene
Basalte, Andesite und deren Tuffe	Quaternary	Quaternary	Quaternary
volcanogenic formations, basic composition	Upper Cretaceous	Upper Cretaceous	non-Holocene
Basalte, Andesite und deren Tuffe	Upper Quaternary	Middle Quaternary	Quaternary
Basalte, Andesite und deren Tuffe	Upper Quaternary	Upper Quaternary	Holocene

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Table S17: Calculated areas of the Kamchatka Peninsula. Note that the Holocene fraction is calculated considering Holocene area, Quaternary area and total area.

Area Holocene (km <sup>2</sup> )	Area Non-Holocene (km <sup>2</sup> )	Area Quaternary (km <sup>2</sup> )	Total Area (km <sup>2</sup> )	Holocene (%)
183.60	55885,588.34	28502,850.52	86228,622.46	2.28

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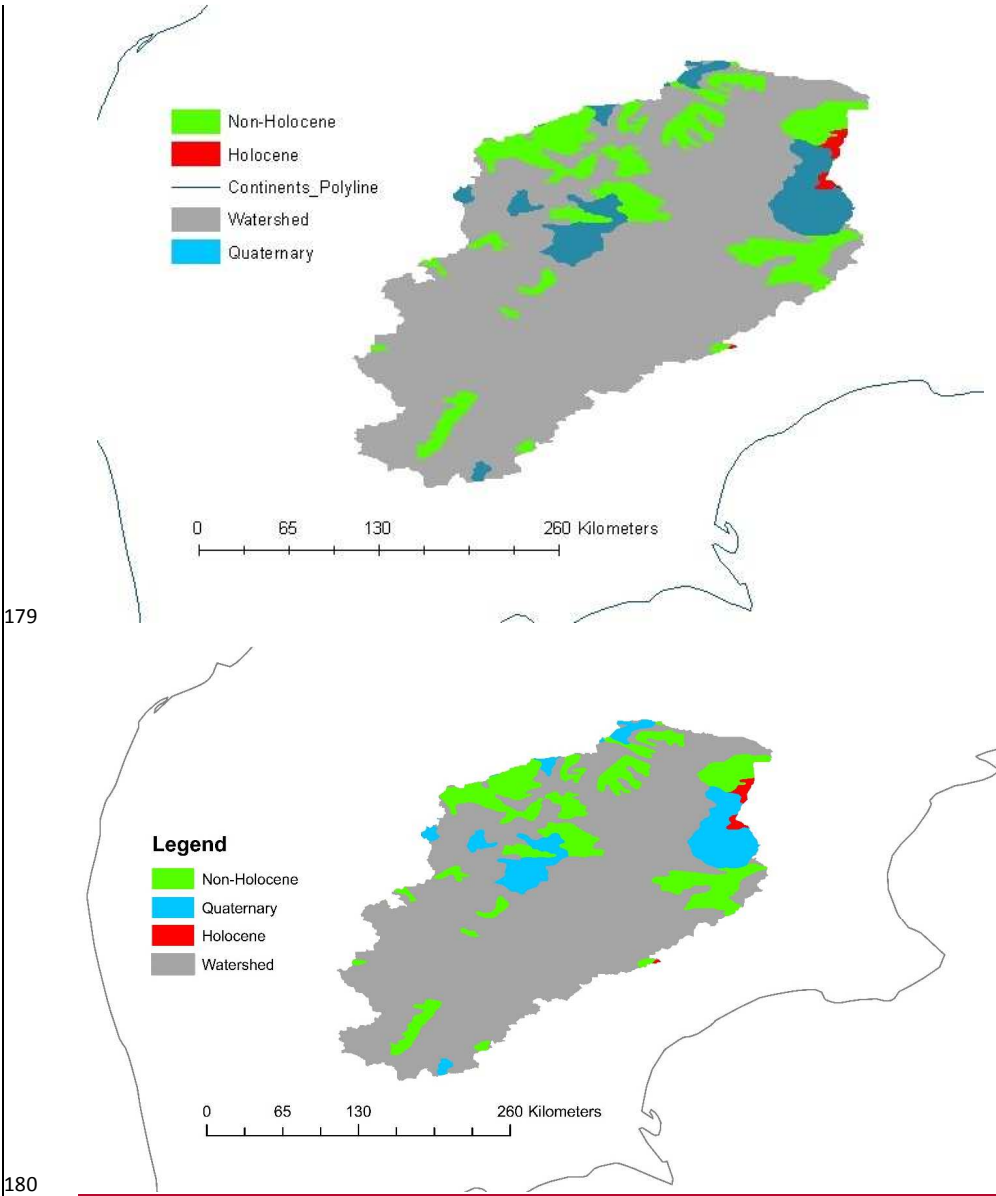


Figure S8: Map showing the watershed of the Kamchatka Peninsula.

|181  
182

183 **Taranaki (No.28)**

184 Taranaki volcano is located in the southern part of the Northern Island of New Zealand.  
185 Price et al. (1992) define four different regions of the Taranaki Volcanics (Paritutu,  
186 Kaitake, Pouakai and Egmont), ~~where Mt. Egmont is the youngest. After Neall et al.~~  
187 ~~(1986) the cone of Mt. Egmont represents about 8% of the total eruptive mass and is of~~  
188 ~~an age of about 10ka (Holocene).with Mt. Egmont as the youngest one. After Price et al.~~  
189 ~~(1992) and references therein the cone of Mt. Egmont represents about 8% of the total~~  
190 ~~eruptive mass and is of an age of about 10 ka (Holocene). Note that Mt. Egmont is not~~  
191 ~~only composed of basalts but also of basaltic andesites and andesites (Price et al., 1992).~~  
192 We assume here a predominantly basaltic composition.

193

194 **Big Island, ~~Hawaii~~Hawaii' (No.29)**

195 The geological map of ~~the~~ Big Island of ~~Hawaii~~Hawaii' was digitized after ~~an original~~a map  
 196 of Stearns and Macdonald (1946). ~~Besides a separation into Holocene and non-Holocene~~  
 197 ~~rocks, two different types of~~The Holocene fraction of the Kilauea and Mauna Loa was  
 198 ~~calculated as~~Quaternary rocks exist:

199 ~~Qu1: Hualalai Volcano: fraction Holocene = Area Qu1 x (11700a / 2580000a)~~

200 ~~Qu2: Kilauea and Mauna Loa Volcano: fraction Holocene = Area Qu2 x (11700a /~~  
 201 ~~426000a)~~Holocene fraction of quaternary areas = Area Quaternary \*  $\left(\frac{11,700}{126,000}\right)$

202 with ~~an~~the assumption ~~of~~that "Latest Pleistocene" = Upper Pleistocene (0.126Ma).

203 ~~In~~The total, ~~a~~ Holocene fraction ~~of 13.73%~~ was 14.08%, calculated ~~by~~:

204  
 205 
$$\text{Holocene fraction} = \frac{\left(\text{Area Holocene} + \text{Area Quaternary} * \left(\frac{11,700}{126,000}\right)\right)}{\text{Total area}}$$

Table S18: Classification of the basaltic rocks of ~~the~~ Big Island of ~~Hawaii~~Hawaii' after Stearns and Macdonald (1946) and our interpretation ("System/Series").

Id	Volcano	Description	Age	System/Series
0	Kohala Mountain	<del>Hawai</del> Hawaii' Volcanic Series, <u>Andesite</u>	Pleistocene	<del>non-Holocene</del> <u>not considered</u>
1	Kohala Mountain	Pololu Volcanic Series	Pliocene and older	non-Holocene
2	Mauna Kea Volcano	Recent lavas	Holocene	Holocene
3	Mauna Kea Volcano	Pleistocene lavas	Pleistocene	non-Holocene
4	Mauna Kea Volcano	Hamakua Volcanic Series, capped by Pahala ash	Pleistocene	non-Holocene
5	Hualalai Volcano	Historic lavas	Holocene	Holocene
6	Hualalai Volcano	Prehistoric lavas, <u>partly younger or older than Waawaa volcanics</u>	Quaternary	<u>non-Holocene</u> <u>Quaternary</u> 1
7	Hualalai Volcano	Waawaa Volcanics (pumice cone, trachyte lava flow, <u>partly covered with</u>	Pleistocene	<del>non-Holocene</del> <u>not considered</u>

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		<u>basaltic lavas and Pahala ash)</u>		
8	Kilauea Volcano	Historic lavas	Holocene	Holocene
9	Kilauea Volcano	Prehistoric lavas	Recent and latest Pleistocene	<del>Quaternary</del> Quaternary
10	Kilauea Volcano	Hilina Volcanic Series, capped by Pahala ash	Pleistocene	non-Holocene
11	Mauna Loa Volcano	Historic lavas	Holocene	Holocene
12	Mauna Loa Volcano	Prehistoric lavas	Recent and latest Pleistocene	<del>Quaternary</del> Quaternary
13	Mauna Loa Volcano	Kahuku Volcanic Series, capped by Pahala ash	Pleistocene	non-Holocene
14	Mauna Loa Volcano	Ninole Volcanic Series	Pliocene or older	non-Holocene

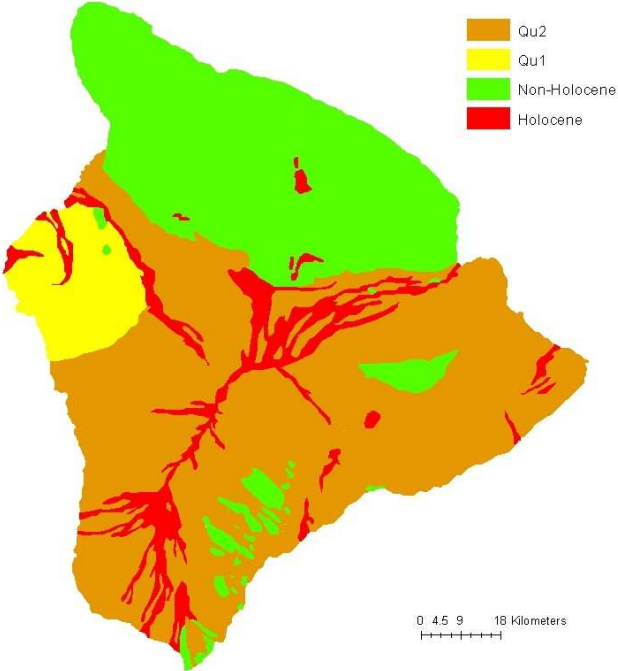
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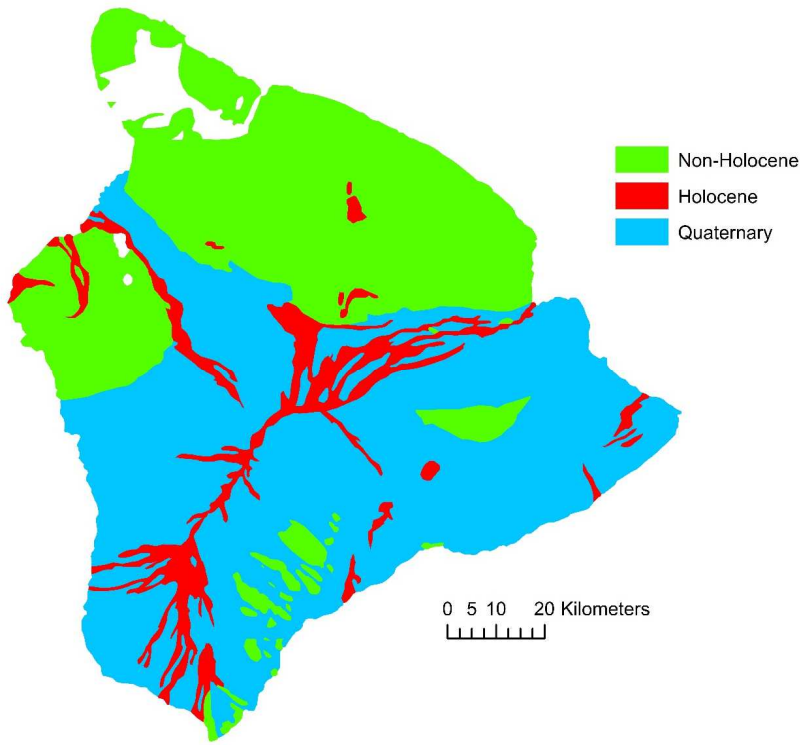
Table S19: Summary of the calculated areas for ~~the Big Island of Hawaii~~Big Island of Hawaii. The Holocene fraction was calculated applying the above mentioned equation considering the Holocene area, Quaternary area and total area.

Area Holocene (km <sup>2</sup> )	Area Non-Holocene (km <sup>2</sup> )	Area Quaternary (km <sup>2</sup> )	Total Area (km <sup>2</sup> )	Holocene (%)
929.17	<del>3357.53</del> 3,786.30	<del>6252.295</del> 5,535.87	<del>40539.09</del> 10,251.34	<del>43.73</del> 14.08

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Figure S9: Map of ~~the~~ Big Island of Hawaii showing the different age distributions of basaltic rocks.

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212 **High Cascades (No.30)**

The geological information for the High Cascades was ~~derived of~~*taken from* the GLiM by Hartmann and Moosdorf (2012) ~~and results in a Holocene fraction of 0.20%.~~

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213 . The Holocene fraction was calculated as for the Virunga province (No.23) and is 0.20%.  
 214 The shapefiles of the watersheds are based on the GLORICH database (Hartmann et al.,  
 215 2014).

Table S20: Classification of the basaltic rocks of the High Cascades Region after the Global Lithological Map, attribute xx='vb' (first three columns) and our interpretation ("System/Series").

Rock_Description	Age_Min	Age_Max	System/Series
basalt;	Early to Middle Miocene	Early to Middle Miocene	non-Holocene
tholeiite; siltstone	Eocene	Eocene	non-Holocene
basalt; andesite	Late Eocene to Oligocene	Late Eocene to Oligocene	non-Holocene
mafic volcanic rock;	Late Miocene to Pliocene	Late Miocene to Pliocene	non-Holocene
basalt; volcanic breccia (agglomerate)	Middle Eocene to Late Eocene	Middle Eocene to Late Eocene	non-Holocene
basalt (tholeiite); andesite	Middle Miocene	Middle Miocene	non-Holocene
basalt;	Middle Miocene	Middle Miocene	non-Holocene
andesite; basalt	Middle to Late Miocene	Middle to Late Miocene	non-Holocene
basalt; andesite	Miocene	Miocene	non-Holocene
basalt; rhyolite	Miocene-Pliocene	Miocene-Pliocene	non-Holocene
basalt; volcanic breccia (agglomerate)	Oligocene to Miocene	Oligocene to Miocene	non-Holocene
basalt; andesite	Pleistocene to Holocene	Pleistocene to Holocene	Quaternary
basalt; andesite	Pliocene to Pleistocene	Pliocene to Pleistocene	non-Holocene
andesite; basalt	Quaternary	Quaternary	Quaternary
andesite; basalt	Tertiary (2-24 Ma)	Tertiary (2-24 Ma)	non-Holocene
Andesite; basalt	Miocene	Miocene	non-Holocene

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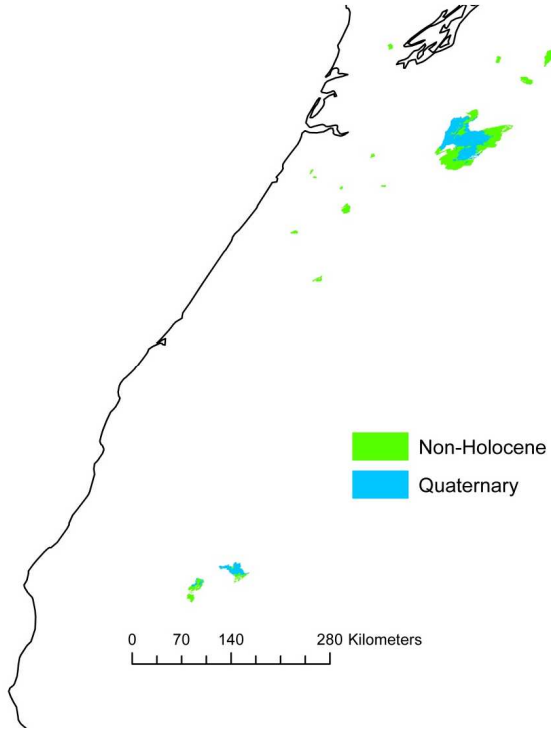
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Table S21: Summarized area calculation for the watersheds of the High Cascades. *The Holocene fraction was calculated as for the Virunga province (No. 23).*

Area Holocene (km <sup>2</sup> )	Area Non-Holocene (km <sup>2</sup> )	Area Quaternary (km <sup>2</sup> )	Total Area (km <sup>2</sup> )	Holocene (%)
0	<del>30333,033.92</del>	<del>24792,479.39</del>	<del>55135,513.31</del>	0.20

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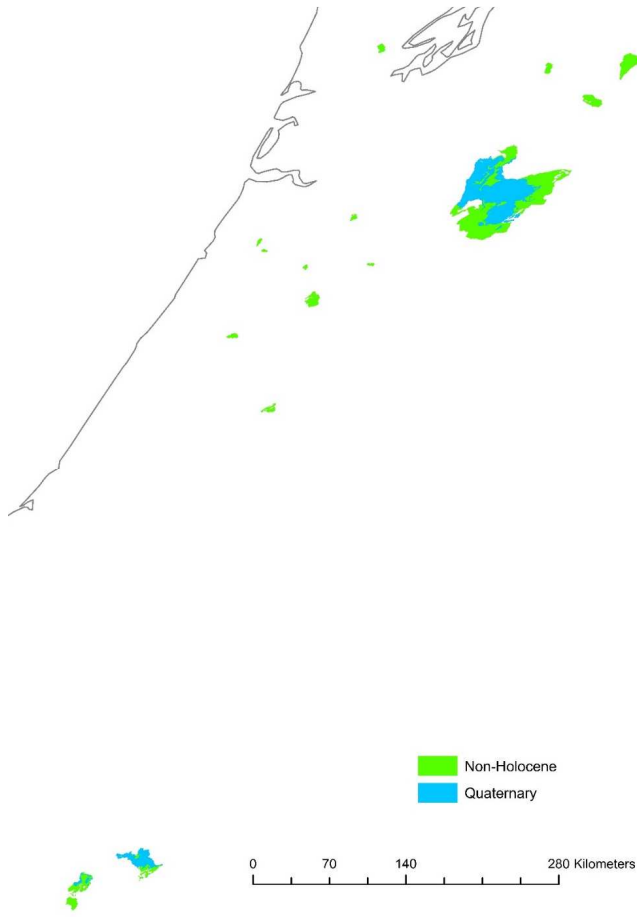


Figure S10: Map of the watersheds of the High Cascades.

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222 **Sao Miguel (No.31)**

223 ~~Sao Miguel is related to a Hot Spot in the northern Atlantic. The geological map was~~  
224 ~~digitized after Moore (1990). There are only two watersheds considered in this study and~~  
225 ~~the age of the basaltic rocks can be divided into two different Quaternary classes:~~

226 Qu1: fraction Holocene = Area-Qu1 x (11700a / 200000a) (Agua de Pau Volcano,  
227 0-200ka)

228 Qu2: fraction Holocene = Area-Qu2 x (11700a / 100000a) (Furnas Volcano, 0-  
229 100ka)

230

*Table S22: Classification of the basaltic rocks of Sao Miguel from the original data and our interpretation ("System/Series").*

<b>Id</b>	<b>Name</b>	<b>Type</b>	<b>Age</b>	<b>System/Series</b>
2	Agua de Pau Volcano	trachyte stratovolcano	0-200ka	Quaternary1
4	Furnas Volcano	trachyte-stratovolcano	0-100ka	Quaternary2

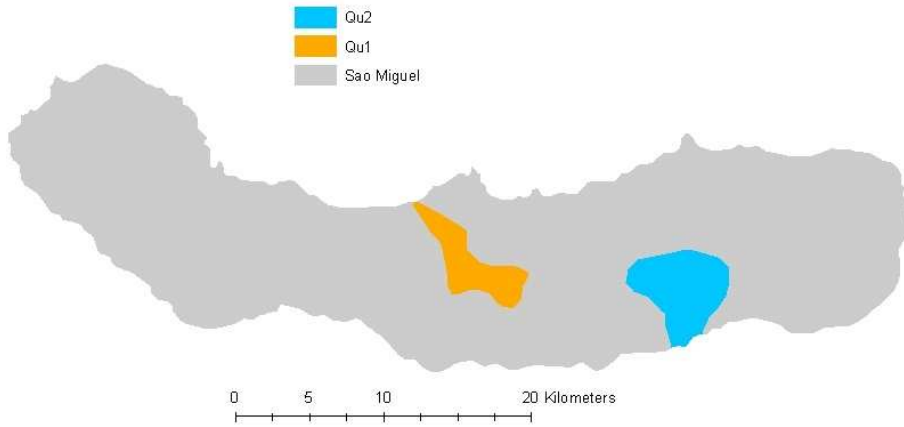
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*Table S23: Summary of the calculated areas of the watersheds of Sao Miguel.*

<b>Area-Holocene (km<sup>2</sup>)</b>	<b>Area-Non-Holocene (km<sup>2</sup>)</b>	<b>Area-Quaternary (km<sup>2</sup>)</b>	<b>Total Area (km<sup>2</sup>)</b>	<b>Holocene (%)</b>
0	0	47.04	47.04	9.35

232



233

*Figure S11: Map of Sao Miguel showing the two watersheds considered in this study.*

234

235 **Iceland (No.3231)**

236 The information of the basaltic rocks of Iceland are ~~derived~~taken from the GLiM  
 237 (Hartmann and Moosdorf, 2012) and ~~can be~~are classified into Holocene, non-Holocene  
 238 and Quaternary (for the calculation see Virunga Province, No. 23) and results in a  
 239 Holocene coverage of 13.19%.

240

Table ~~S24:S22~~: Description of the basaltic rocks of Iceland after the GLiM, attribute  
~~xx='vb'~~ (first three columns) and our interpretation ("System/Series").

Rock Description	Age_Min	Age_Max	System/Series
Basalt and andesite	Holocene (postglacial time)	Holocene (postglacial time)	Holocene
Basic and intermediate hyaloclastites, lavas and associated sediments	Late Pleistocene	Late Pleistocene	non-Holocene
Basic and intermediate lavas and pyroclastic rocks (mainly hyaloclastite)	Late Pliocene, Early Pleistocene, 3.3 -	Late Pliocene, Early Pleistocene, 3.3 -	non-Holocene
Ocean-floor basalt; on land also intermediate volcanic rocks and sedimentary rocks	Miocene	Miocene	non-Holocene
Basic and intermediate volcanic rocks with intercalated sedimentary rocks	Miocene-Early Pliocene, older than 3.3 M	Miocene-Early Pliocene, older than 3.3 M	non-Holocene
Ocean-floor basalt	Pliocene	Pliocene	non-Holocene
Ocean-floor basalt	Quaternary	Quaternary	Quaternary

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Table ~~S25:S23~~: Summary of the area calculation for Iceland. Note that the Holocene  
~~fraction is calculated as for the Virunga province (No. 23).~~

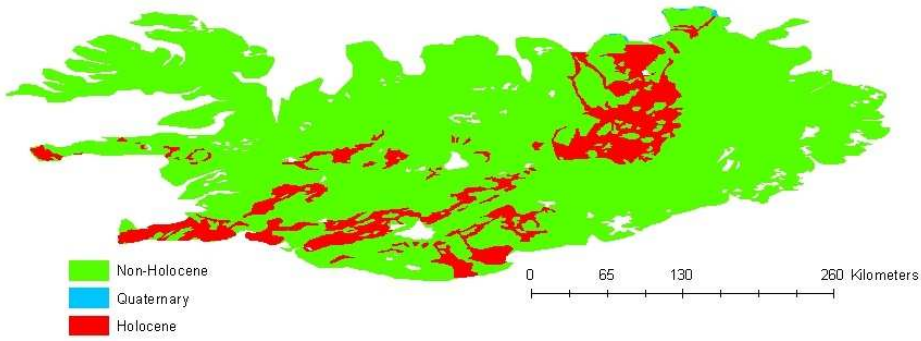
Area Holocene (km <sup>2</sup> )	Area Non- Holocene (km <sup>2</sup> )	Area Quaternary (km <sup>2</sup> )	Total Area (km <sup>2</sup> )	Holocene (%)
<del>13196</del> <u>13,196.86</u>	<del>8677386</del> <u>867,773.61</u>	104.36	<del>100075.00</del> <u>100,074.83</u>	13.19

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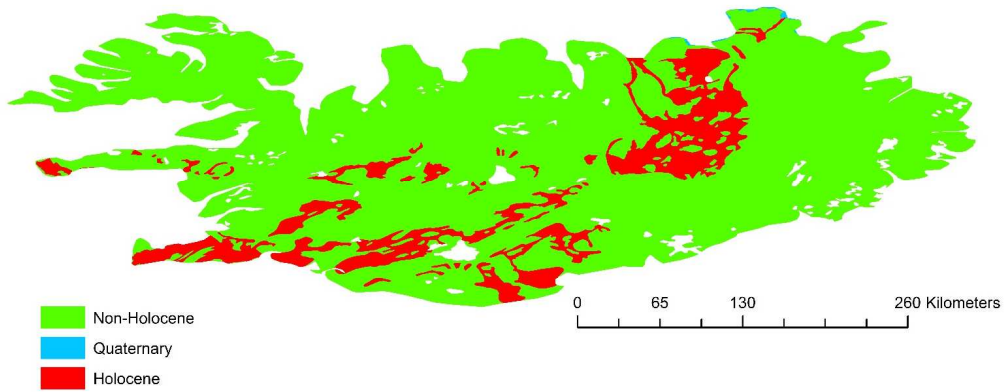


Figure S11:1: Map of Iceland showing the distribution of the age of basaltic rocks.



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259 **Sao Miguel (No.32)**

260 Sao Miguel is related to a Hot Spot in the northern Atlantic. The geological map was  
261 digitized after Moore (1990). There are only two watersheds considered in this study  
262 (Freire et al., 2013). It was assumed that the Holocene area is proportional to the  
263 Holocene time span relative to the given mapped quaternary age:

264 Qu1 (Agua de Pau Volcano, 0 – 200 ka):

265 
$$\text{Holocene fraction} = \text{Area Qu1} * \left(\frac{11,700}{200,000}\right)$$

266 Qu2 (Furnas Volcano, 0 – 100 ka):

267 
$$\text{Holocene fraction} = \text{Area Qu2} * \left(\frac{11,700}{100,000}\right)$$

268 The final Holocene fraction is 9.35% and is calculated by:

269 
$$\text{Holocene fraction} = \frac{\text{Area Qu1} * \left(\frac{11,700}{200,000}\right) + \text{Area Qu2} * \left(\frac{11,700}{100,000}\right)}{\text{Total Area}}$$

270 The watershed areas are of trachytic composition, therefore Sao Miguel was excluded  
271 from the calculations (eq. 5 in the main text) but were kept for comparison because of  
272 the generally basaltic environment of the island.

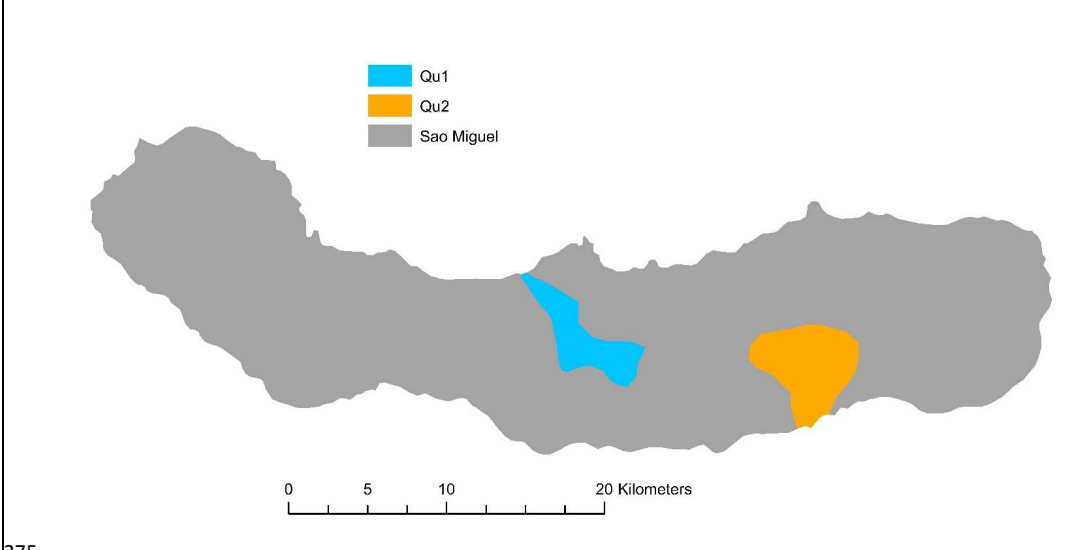
Table S224: Classification of the rocks of the watersheds of Sao Miguel from the original data and our interpretation (“System/Series”).

<b>Id</b>	<b>Name</b>	<b>Type</b>	<b>Age</b>	<b>System/Series</b>
2	Agua de Pau Volcano	trachyte stratovolcano	0-200ka	Qu1
4	Furnas Volcano	trachyte stratovolcano	0-100ka	Qu2

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273 Table S235: Summary of the calculated areas of the watersheds of Sao Miguel. The Holocene fraction was calculated by the above mentioned equation.

<b>Area Holocene (km<sup>2</sup>)</b>	<b>Area Non-Holocene (km<sup>2</sup>)</b>	<b>Area Qu1 (km<sup>2</sup>)</b>	<b>Area Qu2 (km<sup>2</sup>)</b>	<b>Total Area (km<sup>2</sup>)</b>	<b>Holocene (%)</b>
0	0	18.86	28.18	47.04	9.35



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*Figure S12: Map of Sao Miguel showing the two watersheds considered in this study.*

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277 **Tianchi Lake (No.33)**

278 The geological map of the Tianchi Lake region was ~~derived by digitized after~~ Paone and  
 279 Yun (2016). The ~~basalts can be rocks are~~ directly separated into Holocene and non-  
 280 Holocene age. The total ~~basaltic catchment~~ area is 11.31 km<sup>2</sup> and ~~at the~~ Holocene fraction  
 281 area ~~of is~~ 65.34% ~~can be calculated. %~~. The value for the runoff was calculated ~~from after~~  
 282 Fekete et al. (2002).

283

Table S24: Description of the ~~basaltic~~ rocks of Tianchi Lake after Paone and Yun (2016) ~~(first column)~~ and our interpretation ("System/Series").

Description	System/Series
second fan-shaped debris flow - Holocene	Holocene
Rock-fall deposit - Holocene?	Holocene
1668 dark trachytic ignimbrite and surge - Holocene	Holocene
third fan-shaped debris flow - Holocene	Holocene
1903 phreatomagmatic eruption - Holocene	Holocene
Baitoushan III upper trachyte cone with comendite, 0.02-0.22 Ma	non-Holocene
Baithoushan II middle trachyte cone and ignimbrite, 0.25-0.44 Ma	non-Holocene

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284 ~~Since the lithology is mostly described as trachytic, Tianchi Lake was excluded from the~~  
 285 ~~calculations (eq. 5, main text) but kept in this study for comparison.~~

Table S25: Summary of the area calculation for Tianchi Lake. ~~The Holocene fraction~~ was calculated by the ratio of Holocene area/total area.

286

Area Holocene (km <sup>2</sup> )	Area non-Holocene (km <sup>2</sup> )	Area Quaternary (km <sup>2</sup> )	Total Area (km <sup>2</sup> )	Holocene (%)
7.39	3.92	0	11.31	65.34

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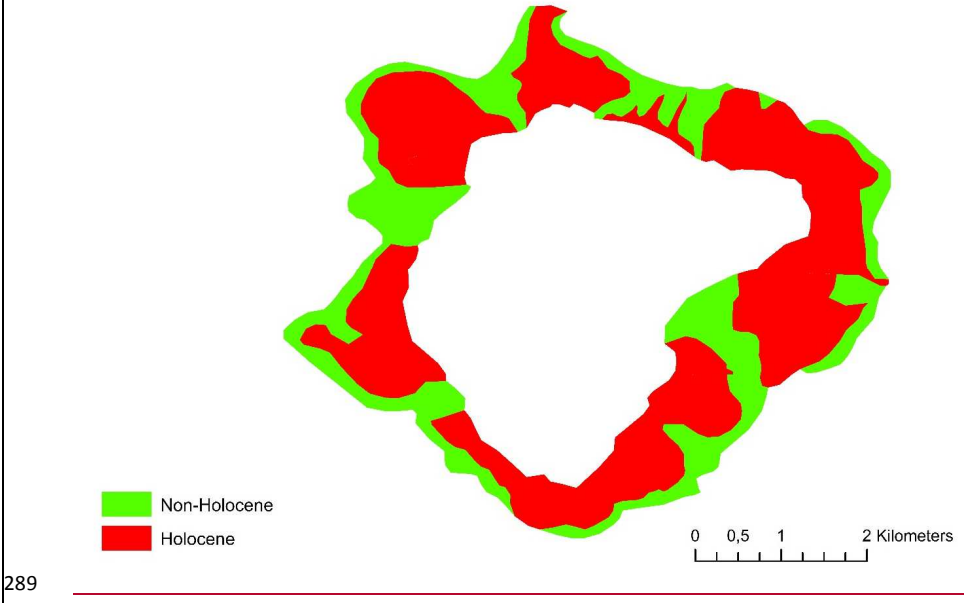
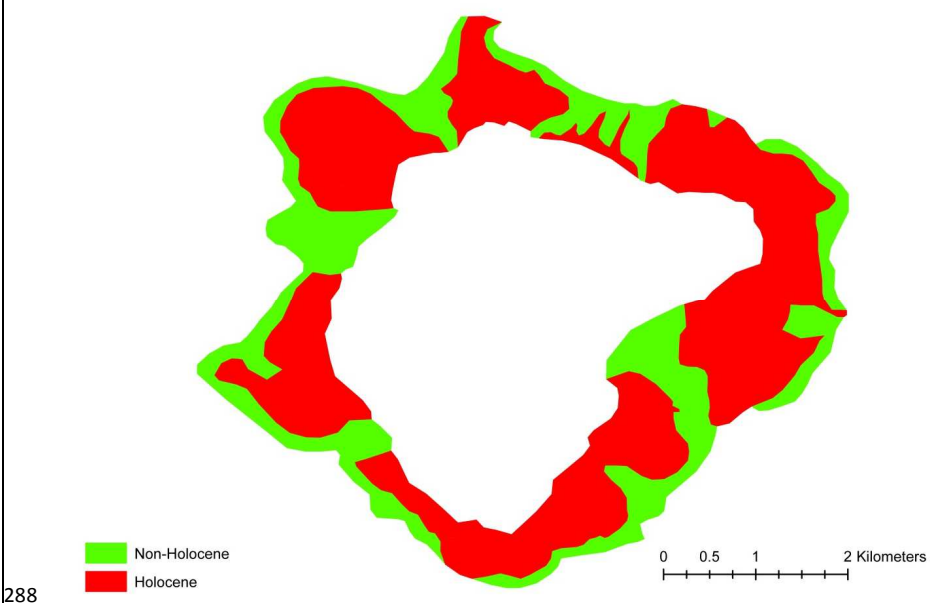


Figure S12: Map of Tianchi Lake and its surrounding lithology showing the distribution of the age of ~~basaltic rocks~~.

Part Two rocks.

290 **Supplemental Material C: Saturation state with respect to calcite**  
291

292 Data used in the previous study of Li et al. (2016) was derived by published literature and  
293 the GLORICH database (Hartmann et al., 2014), but also new river sites of China are  
294 included (for more details see Supplementary Information of Li et al. (2016). Li et al.  
295 (2016) used either alkalinity or DIC concentrations to calculate the alkalinity flux rates of  
296 the individual basaltic volcanic areas. They calculated the alkalinity flux rates by  
297 multiplying the mean concentration of dissolved inorganic carbon (DIC) by the annual  
298 runoff.

299 For six areas full water chemistry data are available (High Cascades, Japan, NE North  
300 America, SE Australia, South Africa and Tasmania), while for others either pH or major  
301 cations were not available. For these available data (103 catchments) the saturation index  
302 for calcite was calculated. In general, water samples are undersaturated with respect to  
303 calcite (77%). From the oversaturated samples 50% have values close to 0 with an  
304 saturation index  $SI < 0.5$ . A  $SI \sim 0.5$  is the typical value for rivers in limestone areas  
305 (Romero-Mujalli et al., 2018). The other 12 values are between  $SI = 0.5$  and  $0.9$  and  
306 according water sample locations are mostly in dry areas of South Africa or Australia.

307 In general, younger active areas have significant contributions of magmatic  $SO_4$  or  $Cl$ ,  
308 which shifts the saturation states normally further to lower, negative values. We cannot  
309 conclude from the river data what happens in the aquifer system but reference the study  
310 of Jacobson et al. (2015), which quantified the contribution of trace calcite dissolution  
311 from basalt using Ca-isotope data.

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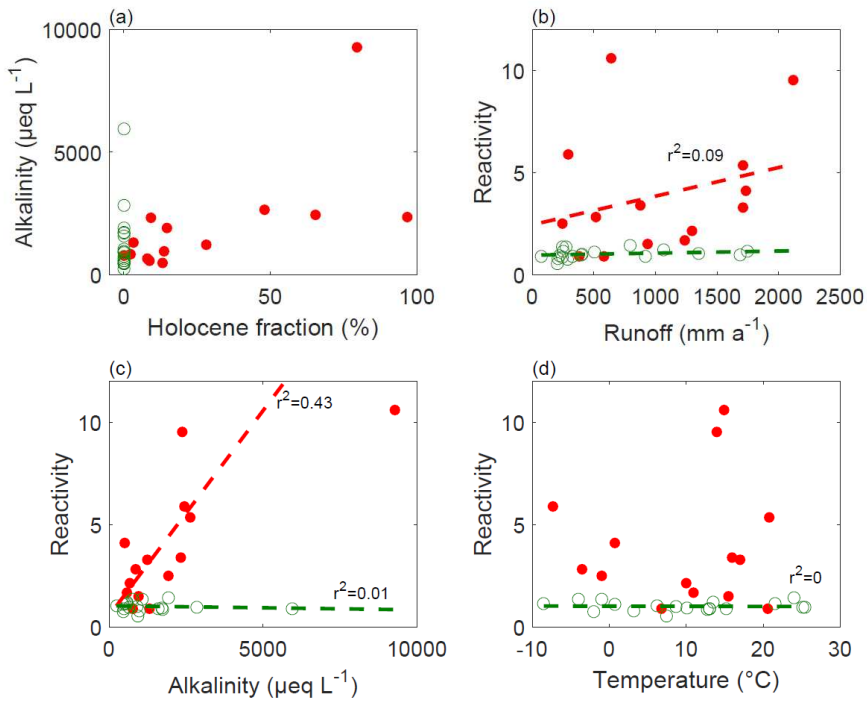
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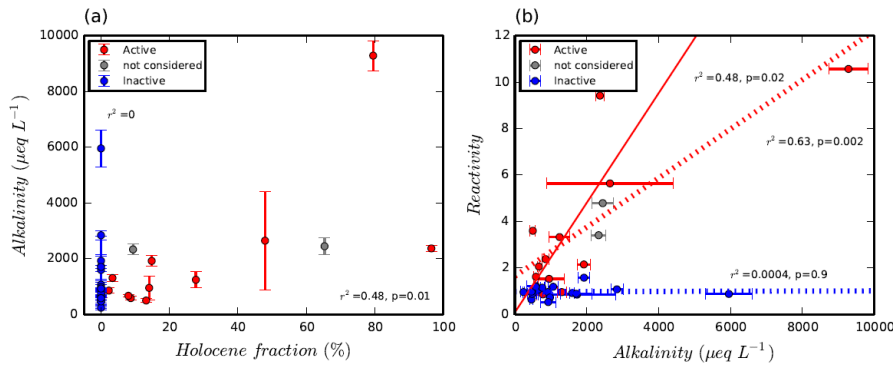
314 **Supplemental Material D: Additional information on relations among studied**  
315 **parameters between Alkalinity, Reactivity and Holocene area fraction**

316 ▲  
317 In addition to results provided in the main text further relationships among applied  
318 parameters are shown.

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322 Figure 14S: Scatterplot relationships between alkalinity, Holocene fraction, reactivity and  
 323 temperature Reactivity. a) Holocene fraction versus alkalinity concentrations. Green  
 324 (blue: IVFs, red: AVFs). b) Reactivity versus runoff. The green regression line (IVFs)  
 325 suggests almost no correlation of reactivity with runoff, also for the AVFs no significant  
 326 correlation is identified (Alkalinity concentration versus Reactivity: For the AVFs the  
 327 coefficient of determination is  $r^2 = 0.09$  and slope of  $63$  ( $p = 0.001$  with reactivity units per  
 328 mm runoff  $\text{a}^{-1}$ ; excluding Mt. Etna and Mt. Cameroon). c) Reactivity versus alkalinity  
 329 concentration: Excluding Mt. Etna, a coefficient of determination  $r^2 = 0.43$  exists,  
 330 suggesting an increase in reactivity due to  $0.02$ ). However, removing the one outlier with  
 331 high alkalinity concentration (Mt. Etna) does not suggest that elevated alkalinity  
 332 concentration for AVFs. d) concentrations are responsible in general for elevated  
 333 Reactivity versus temperature suggests no bias of reactivity due to a temperature effect  
 334 for AVFs, as reactivity is based on a temperature normalized parameterization. For the  
 335 IVFs ( $r^2 = 0$ ) no bias with temperature can be identified due to the good correlation of  
 336 AVFs, considering the alkalinity concentration distribution for IVF. The solid red  
 337 regression line shows the correlation of AVFs excluding Mt. Etna ( $r^2 = 0.48, p = 0.02$ ).

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342 **Supplemental Material E: Estimation of the parameters for the new scaling law**

343 ~~The new scaling law for alkalinity fluxes with land surface temperature (green line) (eq. 5,~~  
344 ~~main text) was derived by a Monte Carlo method simulating 10,000 runs selecting~~  
345 ~~randomly temperature and alkalinity flux rates (assuming a normal distribution) within the~~  
346 ~~range of one standard deviation for each volcanic field;~~

347

~~Table S28: Statistical calculations for the previous linear regression with standard deviations of the parameters. The absolute difference  $D_r$  is calculated by:  $CO_2$~~

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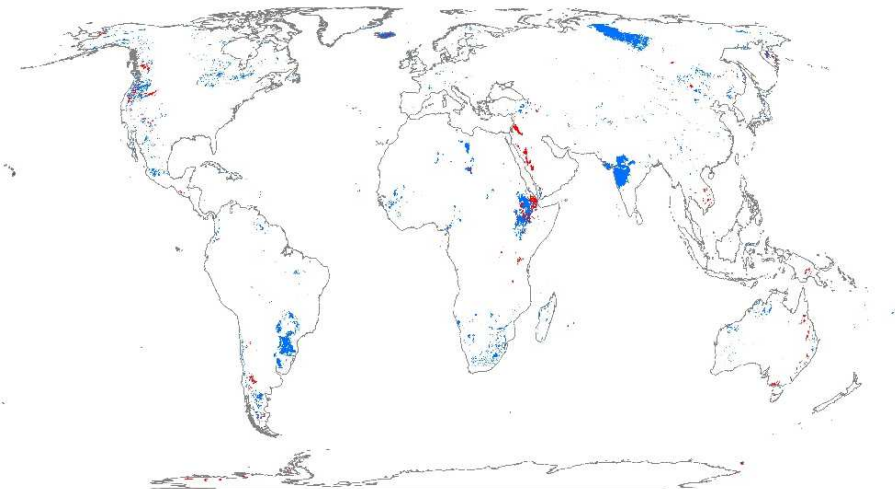
~~consumption<sub>observed</sub> — CO<sub>2</sub> consumption<sub>predicted</sub> and the slope  $x_i - x_{\text{mean}}$  is derived by:  
Holocene fraction — average Holocene fraction.~~

No.	Name	Volcanic activity	CO <sub>2</sub> consumption [10 <sup>6</sup> mol/km <sup>2</sup> /a]		Residuals		Slope	
			observed	predicted	D <sub>r</sub>	D <sub>r</sub> <sup>2</sup>	x <sub>i</sub> -x <sub>mean</sub>	(x <sub>i</sub> -x <sub>mean</sub> ) <sup>2</sup>
1	Massif Central	Inactive	0.372	0.366	0.005	0.000	-11.849	140.401
2	South Africa	Inactive	0.420	0.483	-0.063	0.004	-11.849	140.401
3	Karelia	Inactive	0.131	0.175	-0.044	0.002	-11.849	140.401
4	Coastal Deccan	Inactive	1.110	1.140	-0.030	0.001	-11.849	140.401
5	Interior Deccan	Inactive	1.138	1.164	-0.025	0.001	-11.849	140.401
6	Siberia Traps	Inactive	0.127	0.111	0.016	0.000	-11.849	140.401
7	E'Mei	Inactive	0.321	0.308	0.012	0.000	-11.849	140.401
8	Lei-Qiong	Inactive	1.532	1.056	0.475	0.226	-11.849	140.401
9	Nanjing	Inactive	0.526	0.575	-0.048	0.002	-11.849	140.401
10	Xiaoxinganling	Inactive	0.259	0.187	0.072	0.005	-11.849	140.401
11	Tumen River	Inactive	0.208	0.152	0.056	0.003	-11.849	140.401
12	Mudan River	Inactive	0.204	0.250	-0.046	0.002	-11.849	140.401
13	SE Australia	Inactive	0.441	0.493	-0.053	0.003	-11.849	140.401
14	Tasmania	Inactive	0.377	0.404	-0.027	0.001	-11.849	140.401
15	North Island	Inactive	0.439	0.493	-0.054	0.003	-11.849	140.401
16	Kauai, Hawaii	Inactive	1.026	0.894	0.133	0.018	-11.849	140.401
17	Columbia Plateau	Inactive	0.189	0.335	-0.146	0.021	-11.849	140.401
18	NE America	Inactive	0.235	0.211	0.025	0.001	-11.849	140.401
19	Madeira Island	Inactive	0.618	0.511	0.107	0.011	-11.849	140.401
20	Easter Island	Active	0.757	1.104	-0.347	0.120	-8.489	72.065
21	Mt. Cameroon	Active	5.020	5.431	-0.411	0.169	84.714	7176.452
22	Mt. Etna	Active	5.943	4.861	1.082	1.170	67.711	4584.767
23	Virunga	Active	4.522	4.749	-0.227	0.052	36.171	1308.335
24	La Réunion	Active	2.127	2.408	-0.282	0.079	16.281	265.068
25	Wudalianchi Lake	Active	0.466	0.454	0.013	0.000	2.971	8.826
26	Japan	Active	0.722	0.788	-0.066	0.004	-3.149	9.917
27	Kamchatka	Active	0.444	0.192	0.252	0.064	-9.569	91.568
28	Taranaki	Active	0.864	0.709	0.156	0.024	-3.849	14.816
29	Big Island, Hawaii	Active	0.889	1.354	-0.466	0.217	1.881	3.538
30	High Cascades	Active	0.296	0.328	-0.031	0.001	-11.649	135.701
31	São Miguel	Active	2.047	1.152	0.895	0.801	-2.499	6.245
32	Iceland	Active	0.864	0.477	0.386	0.149	1.341	1.798
33	Tianchi Lake	Active	0.711	0.881	-0.169	0.029	53.491	2861.277
	variance					0.096		0.000
	standard deviation					0.311		0.002

348 **Part Three: Description of the global calculations**

349

350 For the global calculations of alkalinity fluxes, temperature data from Hijmans et al. (2005)  
351 and runoff data from Fekete et al. (2002) were used, as well as a global basalt map,  
352 mostly based on the GLiM (Fig. S15) distinguishing areas considering Holocene area  
353 fractions for the calculations (Cenozoic) and non-Cenozoic basaltic areas.



354

355 
$$X = ((\bar{x} + std) - (\bar{x} - std)) * rand + (\bar{x} - std)$$

356 With X = calculated value for temperature or alkalinity flux rate,

357        $\bar{x}$  = mean value of temperature or alkalinity flux rate,

358       std = standard deviation of temperature or alkalinity flux rate

359 “Rand” is a function from Matlab software which creates uniformly distributed random  
360 numbers (The MathWorks Inc., 2016).

361 The calculated standard deviations of weighted mean values  $\sigma_m$  for temperature and  
362 alkalinity flux rates (Table S1) are taken from Li et al. (2016). The new scaling law is  
363 represented by the equation:

364

365 *Alkalinity flux rate = (1 + b1 \* Holocene fraction) \* b2 \* e<sup>b3\*Temperature</sup>*

366

367 Applying the Levenberg-Marquardt algorithm to the 10,000 sets of alkalinity flux rates and  
368 temperature the following mean b-parameters of the equation were found:

369 b<sub>1</sub> = 0.10 with standard deviation=0.02 and median = 0.10

370 b<sub>2</sub> = 0.23 with standard deviation=0.09 and median = 0.23

371 b<sub>3</sub> = 0.06 with standard deviation=0.02 and median = 0.06

372 The mean residual standard deviation (mrsd) of eq. 5 is 0.3 calculated by the following  
373 equation:

374 
$$mrsd = \sqrt{\frac{\sum(residual)^2}{n-2}}$$

375 Calculating the regression of calculated alkalinity flux rates and estimated alkalinity flux  
376 rates by the new scaling law provides an r<sup>2</sup>=0.96 and p<10<sup>-3</sup>.

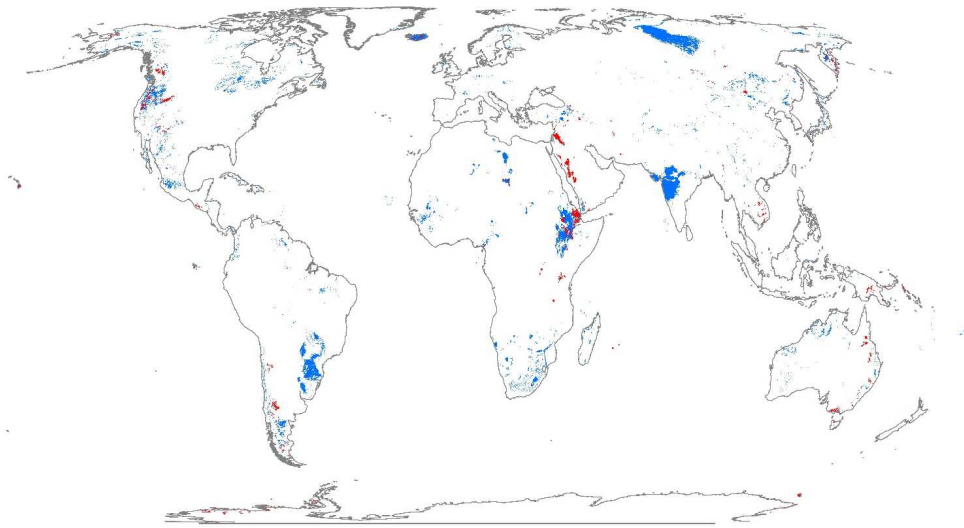
377 The linear regression of Holocene fractions and Reactivity for AVFs gives an r<sup>2</sup>=0.93 and  
378 p=0.01.

379

380 **Supplemental Material F: Global calculations CO<sub>2</sub> consumption by basalt**  
381 **weathering**  
382

383 The calculation of the global CO<sub>2</sub> consumption fluxes by the new scaling law (table S28  
384 and S29, and table 1 in the main text) was done by using the calculated set of 10,000 b-  
385 parameters (Levenberg-Marquardt-algorithm see above, Supplemental Material E) and  
386 global datasets of temperature and runoff (Fekete et al., 2002; Hijmans et al., 2005), as  
387 well as the new global basalt map (Fig. S15). The fluxes were calculated 10,000 times  
388 (according to the set of b-parameters) for each grid cell. Finally, we calculated the mean  
389 values of the fluxes, as well as the standard deviations and the percentiles for 25% and  
390 75% of the global fluxes. This method does not consider the uncertainty due to the  
391 residuals.

392 The global basalt map, mostly based on the GLiM (Fig. S15) distinguishes Holocene,  
393 Cenozoic and non-Cenozoic basaltic areas. The calculations were run within a 20 km grid  
394 resolution.  
395



396  
397 *Figure S15: Global distribution of basalt areas distinguishing Cenozoic (red) and Non-*  
398 *Cenozoic areas (blue). The reported Holocene fractions of the Cenozoic areas are*

399 considered in the global calculation. Some areas with missing age information were  
400 considered as of non-Holocene age and may result in a smaller global Holocene area  
401 fraction ~~that~~ applied for global calculations. The compilation is described in Supplemental  
402 Material G.



403

404 Calculating the global fluxes only once by applying the new scaling law with the mean b-  
405 parameters to every grid cell, and using the global datasets of temperature and runoff  
406 (Fekete et al., 2002; Hijmans et al., 2005), as well as the global basalt map. Here, the  
407 global flux with runoff restriction is  $2 \times 10^{12}$  mol a<sup>-1</sup>. Adding the standard deviation to every  
408 grid cell results in a flux of  $3 \times 10^{12}$  mol a<sup>-1</sup> and subtracting the standard deviation to  
409 every grid cell results in  $1 \times 10^{12}$  mol a<sup>-1</sup>, which is representing the range of uncertainty.

Table ~~S29~~S28: Global calculations of CO<sub>2</sub> consumption for areas with a runoff >74mm/a. As reference previous global empirical equations (Bluth and Kump, 1994; ~~Amiotte-Suchet and Probst, 1995~~; Dessert et al., 2003; Goll et al., 2014; ~~Suchet and Probst, 1995~~) were used to derive CO<sub>2</sub> consumption values were applied to derive global CO<sub>2</sub> consumption values. The

values of the new scaling law in this table and in table S29 are given by simulating 10,000 runs (see above). “p25” and “p75” represent the calculated percentiles of 25% and 75%, respectively and “std” the standard deviation.

	Total Area [km <sup>2</sup> ]	Holocene Area [km <sup>2</sup> ]	% of global basalt area	CO <sub>2</sub> consumption [10 <sup>6</sup> mol/a]					Area [km <sup>2</sup> ] for q>74mm/a	Holocene area [km <sup>2</sup> ] for q>74mm/a
				Goll et al., 2014	Dessert et al., 2003	Amiotte-Suchet & Probst, 1995	Bluth & Kump, 1994	New equation		
Fluxes from purely Holocene areas	55,949	55,949	1.05	17,138	19,388	15,511	11,512	115,206	33,785	33,785
Raster with Holocene influence (Cenozoic)	932,137	55,949	17.46	233,458	247,648	118,919	93,678	314,148	359,190	33,785
Raster without Holocene influence (Non-Cenozoic)	4,406,263	-	82.54	1,237,066	1,319,226	719,451	645,354	1,677,456	3,032,768	-
<b>TOTAL</b>	<b>5,338,400</b>	<b>55,949</b>	<b>100.00</b>	<b>1,470,525</b>	<b>1,566,874</b>	<b>838,371</b>	<b>739,032</b>	<b>1,991,604</b>	<b>3,391,958</b>	<b>33,785</b>

	Total Area [km <sup>2</sup> ]	Holocene Area [km <sup>2</sup> ]	% of global basalt area	CO <sub>2</sub> consumption [10 <sup>6</sup> mol/a]								Area [km <sup>2</sup> ] for q>74mm/a	Holocene area [km <sup>2</sup> ] for q>74mm/a
				Goll et al., 2014	Dessert et al., 2003	Amiotte-Suchet & Probst, 1995	Bluth & Kump, 1994	New scaling law					
								p25	p75	std			
Fluxes from purely Holocene areas	48,593	48,593	0.92	15,531	17,453	13,560	10,060	100,037	89,216	109,234	14,487	29,213	29,213
Raster with Holocene influence (Cenozoic)	903,600	48,593	17.03	225,938	239,122	112,221	88,208	283,726	276,351	292,969	13,074	338,800	29,213
Raster without Holocene influence (Non-Cenozoic)	4,401,200	-	82.97	1,340,243	1,430,216	750,618	658,218	1,646,033	1,586,658	1,708,683	91,215	3,059,600	-
<b>TOTAL</b>	<b>5,304,800</b>	<b>48,593</b>	<b>100.00</b>	<b>1,566,181</b>	<b>1,669,338</b>	<b>862,839</b>	<b>746,427</b>	<b>1,929,759</b>	<b>1,868,357</b>	<b>1,992,574</b>	<b>92,519</b>	<b>3,398,400</b>	<b>29,213</b>

Table S30S29: Global ~~calculations~~CO<sub>2</sub> consumption by basalt weathering without runoff restriction– as applied in Table S28.

	Total Area [km <sup>2</sup> ]	Holocene Area [km <sup>2</sup> ]	% of global basalt area	CO <sub>2</sub> consumption [10 <sup>6</sup> mol/a]				
				Goll et al., 2014	Dessert et al., 2003	Amiotte- Suchet & Probst, 1995	Bluth & Kump, 1994	New equation
Fluxes from purely Holocene areas	55,949	55,949	1.05	17,227	19,484	15,564	11,701	249,201
Raster with Holocene influence (Cenozoic)	932,137	55,949	17.46	234,714	249,063	119,831	96,017	805,917
Raster without Holocene influence (Non-Cenozoic)	4,406,263	-	82.54	1,241,945	1,324,164	722,929	657,897	2,625,217
<b>TOTAL</b>	<b>5,338,400</b>	<b>55,949</b>	<b>100.00</b>	<b>1,476,659</b>	<b>1,573,227</b>	<b>842,760</b>	<b>753,915</b>	<b>3,431,134</b>

412

413

	Total Area [km <sup>2</sup> ]	Holocene Area [km <sup>2</sup> ]	% of global basalt area	CO <sub>2</sub> consumption [10 <sup>6</sup> mol/a]							
				Goll et al., 2014	Dessert et al., 2003	Amiotte- Suchet & Probst, 1995	Bluth & Kump, 1994	New scaling law	p25	p75	std
Fluxes from purely Holocene areas	48,593	48,593	0.92	15,646	17,577	13,628	10,209	209,602	195,275	223,222	19,264
Raster with Holocene influence (Cenozoic)	903,600	48,593	17.03	227,716	241,036	113,265	90,347	735,004	707,519	763,323	38,618
Raster without Holocene influence (Non-Cenozoic)	4,401,200	-	82.97	1,351,814	1,442,505	756,928	670,733	2,534,801	2,434,265	2,646,095	161,340
<b>TOTAL</b>	<b>5,304,800</b>	<b>48,593</b>	<b>100.00</b>	<b>1,579,530</b>	<b>1,683,541</b>	<b>870,193</b>	<b>761,081</b>	<b>3,269,805</b>	<b>3,150,147</b>	<b>3,404,366</b>	<b>193,745</b>

414 **Supplemental Material G: Enhanced global basalt map beyond GLiM**

415 A new global basalt map was ~~derived by the combination of~~ created combining several  
 416 ~~different input~~ datasets. Main input is ~~from~~ the Global Lithological Map (GLiM) (Hartmann  
 417 and Moosdorf, 2012) ~~whereas new~~. New, additional data ~~comes were taken~~ from  
 418 geological maps that were considered for the development of the Global Unconsolidated  
 419 Sediments Map database(GUM) (~~Börker et al., 2018~~)(Börker et al., 2018) and the  
 420 individual active volcanoes described in this study. For the individual calculation of  
 421 Holocene percentages see following tables.

Table ~~S29:S30~~: Table showing the Agessages of the basaltic fields of the GLiM with the authors interpretation (third column). Note that only areas with Holocene fraction are listed. All other age descriptions of the GLiM were considered as of non-Holocene age.

Age_Min	Age_Max	HoloceneArea
Early Pleistocene	Early Pleistocene	0
Himalayan Pleistocene	Himalayan Pleistocene	0
Late Miocene to Pleistocene	Late Miocene to Pleistocene	0
Late Pleistocene	Late Pleistocene	0
Late Pliocene, Early Pleistocene, 3.3-	Late <del>Pliocene</del> , Early Pleistocene, 3.3-	0
Lower Quaternary	Lower Quaternary	0
Lower Quaternary	Miocene	0
Middle Pleistocene	Middle Pleistocene	0
Middle Quaternary	Middle Quaternary	0
Miocene to Pleistocene	Miocene to Pleistocene	0
Mostly Pleistocene	Mostly Pleistocene	0
Neogene	Paleogene	0
Neogene	Neogene	0
Paleogene to Neogene	Paleogene to Neogene	0
Pleistocene	Pliocene	0
Pleistocene	Pleistocene	0
Pleistocene	Late Miocene	0
Pleistocene and Neocene	Pleistocene and Neocene	0
Pliocene to Pleistocene	Pliocene to Pleistocene	0
Quartär, Pleistozän	Quartär, Pleistozän	0
Quaternary (0.15 Ma)	Pleistocene (0.15 Ma)	0
Quaternary (0.23 Ma)	Pleistocene (0.23 Ma)	0
Quaternary (0.26Ma)	<u>Quaternary (0.26Ma)</u>	0
Quaternary (0.37 Ma)	Pleistocene (0.37 Ma)	0
Quaternary (0.64-0.7Ma)	Quaternary (0.64-0.7Ma)	0
Quaternary (0.89 Ma)	Pleistocene (0.89 Ma)	0
Quaternary (1.1 Ma)	Quaternary (1.1 Ma)	0
Quaternary (1.29 Ma)	Quaternary (1.29 Ma)	0
Quaternary, >0.2Ma	Pleistocene, >0.2Ma	0

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Cainozoic	Cainozoic	0.02
Cenozoic	Cenozoic	0.02
Oligocene	Cainozoic	0.02
Quaternary	Tertiary	0.02
Quaternary	Cainozoic	0.02
Quaternary and Tertiary	Quaternary and Tertiary	0.02
QUATERNARY OR TERTIARY	QUATERNARY OR TERTIARY	0.02
Tertiary(?) and Quaternary	Tertiary(?) and Quaternary	0.02
Tertiary-Quaternary	Tertiary-Quaternary	0.02
Oligocene and younger	Oligocene and younger	0.03
Holocene	Miocene	0.05
Miocene to Holocene	Miocene to Holocene	0.05
Miocene to Quaternary	Miocene to Quaternary	0.05
Neogene	Quaternary	0.05
Neogene to Holocene	Neogene to Holocene	0.05
quaternary	neogene	0.05
Quaternary	Neogene	0.05
PLIOCENO-PLEISTOCENO-HOLOCENO	PLIOCENO-PLEISTOCENO-HOLOCENO	0.22
Quaternary	Pliocene	0.22
Quaternary to Pliocene	Quaternary to Pliocene	0.22
Quaternary (0-4 Ma)	Quaternary (0-4 Ma)	0.29
Middle Pliocene to Holocene	Middle Pliocene to Holocene	0.33
Holocene	Pleistocene	0.45
Pleistocene to Holocene	Pleistocene to Holocene	0.45
Quaternary	Upper Quaternary	0.45
Quaternary	Pleistocene	0.45
Quaternary	Quaternary	0.45
Upper Quaternary	Middle Quaternary	0.45
Quaternary, <0.4Ma	Pleistocene, <0.4Ma	2.93
Quaternary (<0.27 Ma)	Pleistocene (<0.27 Ma)	4.33
Late Pleistocene to Holocene	Late Pleistocene to Holocene	9.29
Quaternary (ca. 10-20 000 yrs BP)	Quaternary (ca. 10-20 000 yrs BP)	17
Holocene, <40ka	Holocene, <40ka	29.25
Holocene	Holocene	100
Holocene (postglacial time)	Holocene (postglacial time)	100
Upper Quaternary	Upper Quaternary	100

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424 Table S261: Table showing the age classifications of additional data of several  
 425 geological maps that were used for the GUM database with our interpretation (third  
 426 column). Only the quaternary lithologies were considered. Map sources are:  
 427 Afghanistan (Doeblich et al., 2006), Australia (Raymond et al., 2012), Austria  
 428 (Geologische Bundesanstalt (GBA), 2013), Canada (Fulton, 1995), China (China  
 429 Geological Survey, 2002), Colombia (Gomez Tapias et al., 2015), Ethiopia (Tefera et  
 430 al., 1996), Japan (Geological Survey of Japan AIST (ed.), 2009), Germany  
 431 (Bundesanstalt für Geowissenschaften und Rohstoffe, 2007), Tanzania (Geological  
 432 Survey of Tanzania), USA (Soller et al., 2009).

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433  
 434 Table S30: Table showing the age classifications of additional data of several geological maps  
 435 that were used for the GUM database with our interpretation (third column). Map sources are:  
 436 Afghanistan (Doeblich et al., 2006), Australia (Raymond et al., 2012), Austria (Geologische  
 437 Bundesanstalt (GBA), 2013), Bolivia (GeoBolivia, 2000), Canada (Fulton, 1995), Caribbean  
 438 region (French and Schenk, 2004), China (China Geological Survey, 2002), Colombia (Gomez  
 439 Tapias et al., 2015), Ethiopia (Tefera et al., 1996), Iran (Pollastro et al., 1999), Japan  
 440 (Geological Survey of Japan AIST (ed.) (2009),  
 441 [https://gbank.gsj.jp/seamless/download/downloadIndex\\_e.html](https://gbank.gsj.jp/seamless/download/downloadIndex_e.html), accessed May 2016), North  
 442 Africa 1 (Alimen et al., 1973), North Africa 2 (Alimen et al., 1978), Russia (Zastrozhnev et al.,  
 443 2014), Siegen, Trier, Köln, Frankfurt (Main, west) (Bundesanstalt für Geowissenschaften und  
 444 Rohstoffe, 2007), Spain (Canaries Islands), Spain (Instituto Geológico y Minero de España,  
 445 1988), Tanzania (Geological Survey of Tanzania, Geo-Economic Data (1:2M) — Geology,  
 446 <http://www.gmis.tanzania.com/>, accessed May 2016), USA (Soller et al., 2009).

Age_Min	Age_Max	HoloceneAr ea
0.19M1 M	0.215M26 M	0
2.5 M	0.26 M	0
0.45M215 M	0.65M19 M	0
Sarmatian/PannonianPlio/Pleistocene	Plio/PleistoceneSarmatian/Pannonian	0
PleistocenePliocene	PleistocenePliocene	0
0.26M	4M	0
0.3M	4M	0
0.04M2.5 M	2.5M0.01 M	0
Early Pleistocene	Early Pleistocene	0
0.126780	0.780126	0
Middle Pleistocene	Middle Pleistocene	0
Late Pleistocene	Late Pleistocene	0
Pleistocene	Pleistocene	0
Middle Pleistocene	Late Pleistocene	0
Early Pleistocene	Middle Pleistocene	0

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Cenozoic	Cenozoic	0.02
<del>0</del> 23 Ma	<del>23</del> Ma0 Ma	0.05
<del>Pliocene</del> Holocene	<del>Pliocene</del> Holocene	0.22
Quaternary	Quaternary	0.45
<del>Holocene</del> Late Pleistocene	<del>Late Pleistocene</del> -Holocene	9.29
Holocene	Holocene	<u>100</u>

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