

Response to reviewers:

We thank the two anonymous reviewers for their helpful comments on our manuscript, and provide a response to each of their comments below.

Reviewer 1

This is an interesting test between the possible link of arc volcanism and climate change. It fits the scope of the journal. After the carefully read, I found that the ms has many logical and method flaws, which needs significant revision. The successful link between the arc volcanism and climate change depends on how much carbon dioxide has been outputted through the 40 million years old volcanos. Firstly, the authors claimed there is an intensive eruption pulse at 40 million years based on their own and published data. However, the crucial point is how much 40 Ma volcanism has erupted. The assumption of the authors is improper and geologically impossible.

1-The authors assumed the total area of the 40 Ma is 40,000 km², and the thickness is 3-9km, so the volume of the middle Eocene volcano is 100,000-350,000 km³. 3.9 km is almost the whole thickness of the upper crust, so how could one volcanic eruption make 1/3 of the crust. After I checked the reference Verdel et al., 2011, they claimed the whole Paleogene (66-23 Ma) strata, including the volcanism and sedimentary rocks in the UDMA is 3-9 km. Clearly, the authors have much overestimated the thickness of 40 Ma volcanic rocks. According to figure 2, we see volcanic events throughout the whole Eocene. Although there is an intense event at 40 Ma, still, the 40 Ma volcanic rocks are only a part of the Paleogene volcanic strata (3-9 km). You must be precise how thick is the 40 Ma rock.

The key point of our response to this comment is that there is no one single eruption around 40 Ma. This was perhaps not stated clearly enough in the manuscript, so we have put more emphasis on this. We do not state that all these volcanic rocks have erupted in a single volcanic eruption as the reviewer seems to imply. Rather, we see a large increase in volcanic activity all around Iran in the middle Eocene, and this activity is observed in all different regions. Consequently, there must have been many volcanic eruptions that all together contributed to the thickness of (middle) Eocene volcanic units in Iran. We further emphasise that the thicknesses that we report are fully in line with the statement of Verdel et al. (2011): "Reported thicknesses of Paleogene volcanic and sedimentary rocks are ~3–9 km in the Urumieh-Dokhtar belt (Figure 1) in central Iran and the Alborz Mountains in northern Iran [e.g., Förster et al., 1972; Annells et al., 1975; Hassanzadeh, 1993; Morley et al., 2009]." This is also clear from other literature on this topic. Berberian & King (1981) state that "Extensive volcanism, with a wide range of composition, started in the Eocene Period (50 Ma) and continued for the rest of the period with the climax in Middle Eocene time (about 47-42 Ma). Despite their great thickness (locally up to 6 and 12 km) and wide distribution, the volcanics and tuffs were formed within a relatively short time interval." Stöcklin (1974) mentions Eocene volcanic rocks in the Alborz to have a thickness of 3-5 km. Allen et al. (2003) mention a thickness of 5 km for the Eocene Karaj Formation (which consists mostly of volcanic and volcanoclastic rocks) in the Alborz. Taking into account all these different estimates, we feel that the values of 3-9 km that we use in our calculations are reasonable and agree well with estimates from literature.

To clarify this issue in the text, we have added the above references to support the statement on the thickness of the volcanic deposits. We have modified line 136-139 to: "In the Alborz and Central Iran, middle Eocene extrusive volcanic formations are reported to be very thick, with estimates ranging from 3-5 km in the Alborz Mountains (Stöcklin, 1974), to 6-12 km locally throughout nearly all of Iran (Berberian and King, 1981). More recent estimates of the thickness are 3-9 kilometers (e.g. Morley et al., 2009; Verdel et al., 2011)."

Moreover, we have clarified that we do not intend to suggest that one eruption caused all these deposits but that they rather represent a phase of intensified volcanism, by adding (line 136): “These volcanic rocks were produced by numerous eruptions throughout the middle Eocene.”

2-The second point is that the authors probably underestimated CO₂ output based on their calculation. The authors compared the size of the arc volcanism with the large igneous province in Deccan and directly used the CO₂ output data from LIPs. However, the compositions of arc volcanism are fundamentally different from those of LIP. The arc volcanism is more felsic that is compared to the dominated basalt of LIPs. Then the arc volcanism is much enriched with volatile like carbon (0.6-1.3 wt%, Wallace et al.,2005), water (4 wt%, Plank et al.,2013.). Therefore, if the authors used the arc data, I think the output of carbon maybe more. Because of the compositional difference, the felsic arc volcanism is more like to interact with the carbonate to form skarn that further releases more CO₂. The LIP basalts are more likely to assimilate with carbonate and related to fewer CO₂ (Carter et al., 2016). On the contrary, the basalts are much easier to weathering, which consumes many CO₂, which may cause cooling.

We fully agree with the reviewer that our estimates are likely underestimates. We deliberately chose a conservative approach. Therefore, we mention in line 139-140 that our assumption of a similar volume versus emission relationship as the Deccan traps results in a minimum estimate of CO₂. We will put more emphasis on our estimates being minimum estimates, and the carbon contribution by Eocene volcanism in Iran could have been much larger (see also our response to the next comment).

We have added the following part to the discussion (lines 171-175): “Erosion has affected the entire Iranian plateau, and could have eroded away significant volumes of Eocene volcanic rocks. Morley et al. (2009) and Ballato et al. (2011) note that clasts in the Lower and Upper Red formation (Oligocene-Miocene age), which in many places overlie Eocene volcanics, are for a large part made up of eroded Eocene volcanic rocks. Original thicknesses of Eocene volcanic rocks in Iran could thus have been larger, making our CO₂ output estimate a minimum estimate.”

3-Current data do not support their conclusion. The authors must recalculate the budget. MECO is a global effect. I suggest the authors also add some discussion on the possible Eocene arc volcanism at other places like along the Tethyan region and the Cordillera region in the eastern Pacific. As far as I know, the post-Laramide volcanism is also very strong.

We agree with the reviewer that other volcanically active areas in the Tethyan region might have played a role during the MECO (Armenia, Georgia and Turkey) as we have mentioned in lines 46-48 (in revised manuscript lines 54-56). In addition, we have added the following part to the discussion (lines 141-145): “Our estimates of CO₂ release due to middle Eocene volcanism in Iran are likely underestimates, as there is volcanism in other regions along the Neotethys subduction zone. Unfortunately, the lack of shapefiles of Eocene volcanic and intrusive rocks in Armenia and Azerbaijan, along the Lesser Caucasus Mountains (e.g. Allen and Armstrong, 2008), and plutons and volcanic rocks in Armenia (e.g. Moritz et al., 2016; Sahakyan et al., 2016), hampers calculations on additional CO₂ emissions within these regions.”

We have done a thorough literature study of other inferred causes of the MECO around the globe, such as increased mid-ocean ridge volcanism (Bohaty et al., 2009), increased metamorphic decarbonation associated with Himalayan uplift (Kerrick and Caldeira, 1999; Pearson, 2010), increased extrusive arc volcanism in the Pacific rim (Cambray and Cadet, 1996), increased carbonatite magmatism in the East African Rift (Bailey, 1992, 1993), or increased Cordilleran belt volcanism (Kerrick and Caldeira, 1998). However, we did not find confirming radiometric ages or

other evidence that indicated that other regions showed a temporal link to the MECO event. We thus decided to focus on our own data, instead of discussing the absence of evidence from other regions.

Reviewer 2

Detailed comments on the manuscript of “Exploring a link between the Middle Eocene Climatic Optimum and Neotethys continental arc flare-up” have been made as follows. This paper presents new data, idea and explanation about a link between the Middle Eocene Climatic Optimum and Neotethys continental arc flare-up. It is sure that this interpretation in this paper presented will therefore be of considerable helpful for anyone working in this field. I fully support publication of this work, and the comments that I have listed below are chiefly intended to help the authors make their manuscript as clear and accessible to potential readers as possible.

We thank the reviewer for their kind words and support of our manuscript.

I suggest that the author may consider adding a new section of “Geological background”. The authors may briefly review all previous studies and ideas partly concerning with the relation between petrogenesis and tectonic evolution history based on clearly and strongly geological evidence because conflicting data and hypotheses concerning about geological history and petrogenesis in the studied area have been presented in previous studies. I think that if there is the description about the geological outline, which is also ok although it seems a little simple.

We agree with the reviewer there are many different and conflicting tectonic and petrogenic models for Eocene volcanism in Iran. A thorough review of all of the geologic settings of these different areas of Iran is a huge task that is deserving of a study in its own right. We mainly intend to show in this study that there is a huge increase in volcanism in Iran during the Eocene in all of these regions, regardless of their tectonic history and petrogenesis, which is why we do not discuss all the petrologic models in detail. To give some more background information, we have added to the Introduction (lines 46-53):

“Geologic settings of the Eocene volcanic regions in Iran differ. Extensive magmatism in the Lut block is regarded by Pang et al. (2013) to be the result of post-collisional convective removal of the lithosphere and not directly related to subduction. Volcanism in the Sabzevar zone is linked by Moghadam et al. (2016) to lithospheric delamination, possibly assisted by slab-breakoff. In the Talesh/Alborz region, there are conflicting theories on the formation of the volcanic rocks. Asiabanha & Foden (2012) mention a post-collisional transition to a continental arc in their title, but then describe the volcanism as back-arc volcanism. Van der Boon (2017) gives an overview of proposed conflicting settings for volcanism in the Alborz. It is striking that in most of the areas in Iran, the flare-up is linked to an extensional setting (e.g. Verdel et al., 2011), which makes it different from other flare-ups (e.g. Ducea et al., 2015; Ducea and Barton, 2007).”

Importantly, magmatism (including volcanism) with different characteristics in geochemical composition, mantle source regions and geodynamic setting would have full differences in eruptive column heights for volcanism only, volatile (including CO₂) degassing rates and fluxes, and amounts of outgassing gases from magmatic activities, which are importantly controlling parameters on climate changes related to magmatism (including volcanism). If calculated and/or analysed results of the parameters (including the eruptive column heights for volcanism only, volatile (including CO₂) degassing rates and fluxes, and amounts of outgassing gases from magmatic activities) cannot be well determined by the magmatic (including volcanic) bodies themselves based on the melt inclusion sample analysis in the lab (Including EMP, Raman, SIMES, etc.), instead of comparison with those released from other volcanic activities (e.g. the Deccan traps in this paper), the final results and even conclusions of which would possibly need to reevaluated, because it is not easy to develop a link in these parameters (including the eruptive column heights for volcanism only, volatile (including CO₂))

degassing rates and fluxes, and amounts of outgassing gases from magmatic activities) between magmatism (including volcanism) with different characteristics in geochemical composition, mantle source regions and geodynamic setting. I suggest that the author may further explain the petrologic reason, rationale and geochemical basis of the comparisons in magmatic CO₂ outgassing rate (or amount) between the Deccan traps and magmatic activities in this paper (see details in about Line 140), which may be thought to be an potentially estimated method of the magmatic CO₂ outgassing rate (or amount).

We fully agree with the reviewer that more detailed research on this topic could strengthen or invalidate our results, and we hope that our study encourages further study on the Iranian Eocene volcanics and their CO₂ emissions. Here we describe the state-of-the-art regarding the dating of the volcanic deposits. There is currently not a lot of data available on Eocene melt inclusions in Iran, there are only very few that are focused on mineralisation, so this kind of work could provide more insights into settings of Eocene volcanism, ideally on a similar large scale as we present our dating.

In order to bridge the gap between the scales, we thus have to rely on the scarce information that is available on magmatic volumes and related CO₂ content, and only the well-studied Deccan traps have estimates for this. We thus use what is available, and that is unfortunately only information from the Deccan traps. To our knowledge, there have been no studies that constrained the amount of CO₂ per volume of arc volcanic rocks. We note that that is also a more difficult task, due to the varied nature of the different rock types in arcs (i.e. nearly every type from mafic to felsic, while LIPs consist mainly of basalt).

To comply with the reviewer's comment, we have modified lines 132-133 (in revised manuscript lines 146-147) to: "Due to the absence of quantifications of the relation between the erupted volumes of volcanic rocks and emission of CO₂ in continental arcs, we make a comparison with the Deccan traps, for which this relation has been calculated." As mentioned in lines 139-140 (in revised manuscript lines 153-154), this likely results in a minimum estimate for the amount of CO₂ related to Eocene volcanic activity in Iran.

Additionally, it should really be pointed out here that magmatism concerned with in this paper belongs to HKCA volcanism, which is related to oceanic plate subduction. But, many previous studies (including a recent study published in Geology-2019) indicate that this kind of HKCA volcanism may act as a key driver of the late Paleozoic ice age (Soreghan, G.S., Soreghan, M.J., and Heavens, N.G., 2019, Explosive volcanism as a key driver of the late Paleozoic ice age: Geology.). Thus, magmatism with similar geodynamic setting may have total different the magmatic CO₂ outgassing rate (or amount), which are very comment situations.

The study of Soreghan et al. 2019 is very intriguing but at the same time highly speculative. For example, Lee and Dee (2019) discuss the Soreghan et al. paper, and state that individual eruptions might manifest as short-term cooling events superimposed on an otherwise warmer baseline. This is more consistent with the paradigm.

The Eocene in Iran consists of many units that contain volcanoclastic rocks that have been interpreted as the result of explosive eruptions that might potentially cause some degree of dimming (e.g. Asiabanha et al., 2012; Asiabanha and Bardintzeff, 2013). Many of the Eocene volcanic units in Iran are mapped as 'Eocene volcanics' and which doesn't allow us to precisely quantify the amount of pyroclastics and ignimbrites, as Soreghan et al. (2019) have done. Also eruption magnitudes are not estimated for Eocene volcanic rocks in Iran, and there have been no reports of large calderas besides one in Tafresh (Ghorbani & Bezenjani, 2011).

Most importantly, however, we here test for a link between a phase of global warming through volcanic CO₂ forcing rather than a cooling through volcanic aerosol formation. For these reasons, we at this point choose not to discuss this issue.

However, the Deccan traps and magmatic activities in this paper have totally different geodynamic settings, thus i hope the author may further explain the reason of the comparisons in magmatic CO₂ outgassing rate (or amount) between the Deccan traps and magmatic activities in this paper (see details in about Line 140). Whether or not are the results from the comparisons in this paper better than those in previous studies (Including EMP, Raman, SIMES, etc.)?

Please see our response to a similar comment above.

Other changes:

- In Figure 3, we moved the position of the labels of the peaks (e.g. Peak at 29.5 Ma) slightly to improve readability as it was overlapping with the graph in the first version.
- Removed 'extrusive' in line 18,136 and 138 as that is a pleonasm when combined with 'volcanic'.
- Added reference Boscolo-Galazzo et al. 2013 in the introduction (line 28).

Exploring a link between the Middle Eocene Climatic Optimum and Neotethys continental arc flare-up

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15 **Abstract.** The Middle Eocene Climatic Optimum (MECO), a ~500 kyr episode of global warming that initiated at ~40.5 Ma, is postulated to be driven by a net increase in volcanic carbon input, but a direct source has not been identified. Here we show, based on new and previously published radiometric ages of volcanic rocks, that the interval spanning the MECO corresponds to a massive increase in continental arc volcanism in Iran and Azerbaijan. Ages of Eocene volcanic rocks in all volcanic provinces in Iran cluster around 40 Ma, very close to the peak warming phase of the MECO. Based on the spatial extent and volume of the volcanic rocks as well as the carbonaceous lithology in which they are emplaced, we estimate the total amount of CO₂ that could have been released at this time corresponds to between 1500 and 11300 Pg carbon. This is compatible with the estimated carbon release during the MECO. Although the uncertainty in both individual ages, and the spread in the compilation of ages, is larger than the duration of the MECO, a flare-up in Neotethys subduction zone volcanism represents a plausible excess carbon source responsible for MECO warming.

Deleted: extrusive

25 **1 Introduction**

The MECO is characterized by surface and deep ocean warming, both of approximately 2-6°C. MECO warming initiated at ~40.5 Ma, culminating in a short peak warming phase at ~40.0 Ma and terminating at ~39.9 Ma with a comparatively rapid cooling (Bijl et al., 2010; Bohaty et al., 2009; Bohaty and Zachos, 2003; Boscolo Galazzo et al., 2013, 2014; Cramwinckel et al., 2018). The MECO is associated with a rise in atmospheric CO₂ concentrations (Bijl et al., 2010; Henehan et al., 2020), extensive deep sea carbonate dissolution (Bohaty et al., 2009) and marine biotic change (Bijl et al., 2010; Cramwinckel et al., 2019; Edgar et al., 2013; Witkowski et al., 2012). The MECO inherently differs from the early Paleogene transient

warming events such as the Paleocene-Eocene Thermal Maximum (PETM; ~56 Ma) primarily in its longer duration (~500 kyr) of warming, precluding a sudden trigger but rather suggesting a continued driver (Bohaty and Zachos, 2003; Sluijs et al., 2013). Furthermore, unlike the PETM and similar transients, the MECO is not characterized by a negative $\delta^{13}\text{C}$ excursion of the exogenic carbon pool, ruling out the input of ^{13}C -depleted organic-sourced carbon as a driver, but suggesting a volcanic source (Bohaty and Zachos, 2003). Reconstructions and simulations of the carbon cycle indeed point to an imbalance in the long-term inorganic carbon cycle during the MECO (Sluijs et al., 2013), caused by enhanced volcanism and sustained by diminished continental silicate weathering (van der Ploeg et al., 2018). However, this scenario is quantitatively far from settled, partly because recent analyses based on foraminifer boron isotope ratios suggest that atmospheric CO_2 concentrations rose by significantly less than a doubling and did not rise substantially during the onset of the MECO (Henehan et al., 2020). In addition, a plausible source of excess volcanic CO_2 remains to be identified.

Here, we explore a volcanic arc flare-up in the Neotethys subduction zone as a potential source. Arc flare-ups can generate 80-90% of the total volume of igneous rocks in arc systems in periods of a few million years (Ducea and Barton, 2007). During the Eocene, a large flare-up took place in vast areas of present-day Iran (see Figure 1A) and these volcanic rocks show subduction-related geochemical signatures, representative of continental arc volcanism (Moghadam et al., 2015; Pang et al., 2013; Verdel et al., 2011). Geologic settings of the Eocene volcanic regions in Iran differ. Extensive magmatism in the Lut block is regarded by Pang et al. (2013) to be the result of post-collisional convective removal of the lithosphere and not directly related to subduction. Volcanism in the Sabzevar zone is linked by Moghadam et al. (2016) to lithospheric delamination, possibly assisted by slab-breakoff. In the Talesh/Alborz region, there are conflicting theories on the formation of the volcanic rocks. Asiabanha & Foden (2012) mention a post-collisional transition to a continental arc in their title, but then describe the volcanism as back-arc volcanism. Van der Boon (2017) gives an overview of proposed conflicting settings for volcanism in the Alborz. It is striking that in most of the areas in Iran, the flare-up is linked to an extensional setting (e.g. Verdel et al., 2011), which makes it different from other flare-ups (e.g. Ducea et al., 2015; Ducea and Barton, 2007).

The main volcanic arc associated with the Neotethys subduction zone stretches from Bazman in southeast Iran towards Azerbaijan in the northwest, from where it continues westwards into Armenia, Georgia and Turkey (van der Boon et al., 2017). North of the volcanic arc, in the Peri-Tethys basin of Azerbaijan and Russia, thick bentonites and ash layers are found within middle Eocene marine sediments (Beniamovski et al., 2003; Seidov and Alizade, 1966).

Sahandi et al. (2014) produced a compilation of geological maps of Iran, which shows that more than half of the outcrop area of igneous rocks in Iran is of Eocene age (see Figure 1A). The total surface area that is covered by Eocene igneous rocks is almost 70.000 km^2 (including units mapped as Middle Eocene, Eocene-Oligocene, etc.). A causal relationship between peak volcanism in this region and the MECO has been suggested (Allen and Armstrong, 2008; Kargaranbafghi and Neubauer, 2018), but radio-isotopic age constraints to test this hypothesis are insufficient. To quantitatively assess whether volcanism in the Iran-Azerbaijan region could have been a contributor to global warming during the MECO, we present a compilation of new and previously published radiometric ages for volcanic rocks and estimate eruptive volumes of the flare-up in Iran to evaluate how much CO_2 could have been released during this event.

2 Dating the continental arc flare-up of the Neotethys subduction zone

2.1 New $^{40}\text{Ar}/^{39}\text{Ar}$ data

70 We analyzed 48 samples of Eocene volcanic rocks of the Azerbaijan-Bazman Arc in Iran and Azerbaijan. Lava flows of the Peshtasar formation were dated by Vincent et al. (2005) and van der Boon et al. (2017), but ages suffered from severe excess argon. Here, we re-dated lava flows from the lower and middle part of the Peshtasar formation using new instrumentation to check for potential age bias caused by hydrocarbon interferences in previous data. We further dated samples of two ash layers in the Kura basin in Azerbaijan, as well as four volcanic rocks from the Talesh and western Alborz in Iran (see Figure 75 1B). Depending on the rock type, groundmass, plagioclase, sanidine, biotite and/or glass was measured (see Table 1). Thin section analysis showed pervasive alteration of volcanic rocks, disqualifying many sampled units for radio-isotope dating (see supplementary file S1 for a comparison of some thin sections). However, 8 samples showed no significant alteration and were prepared for $^{40}\text{Ar}/^{39}\text{Ar}$ dating using standard mineral separation techniques including heavy liquid and magnetic separation and handpicking. In general, fractions between 250-500 μm size were taken. For some minerals, both groundmass 80 or glass and plagioclase or biotite could be separated.

Samples were leached with diluted HNO_3 and/or HF. Samples were irradiated during resp. 12 and 18 hours in two irradiations (VU101 in 2014 and VU107 in 2016) at the Oregon State University Triga CLICIT facility, together with Fish Canyon Tuff sanidine as standard (FCs; 28.201 ± 0.023 Ma; Kuiper et al., 2008). After irradiation samples were loaded in Cu-trays and run on a 10-collector Helix-MC mass spectrometer with an in-house built extraction with SEAS NP10, St172 85 and Ti sponge getters and a Lauda cooler run at -70°C , at the Vrije Universiteit Amsterdam. The used cup-configuration was either ^{40}Ar on the H2 Faraday cup and 39-36 argon isotopes on compact discrete dynodes, or both ^{40}Ar and ^{39}Ar on respectively H2 and H1 Faraday. Gain calibration was done by peakjumping CO_2 in dynamic mode on the different cups (see Monster, 2016 for details). Samples were analyzed using step-heating experiments, while for the ash layers usually single or a few grains were fused in one step and analyzed. Initial measurements were on single or a small number of grains, leading 90 in some samples to very low intensities of ^{40}Ar (3-4 times higher than blanks). In those cases, more grains were loaded in the next experiment. Ages are calculated relative to the age of FCs reported in Kuiper et al. (2008; 28.201 ± 0.023 Ma) with decay constants of Min et al. (2000).

Out of the 8 prepared samples, 7 gave results. Our new $^{40}\text{Ar}/^{39}\text{Ar}$ ages from igneous rocks and ash layers fall within a range of ~ 36 -45 Ma (Figure 2A), with weighted mean ages per sample between 39.3-43.1 Ma (Figure 2B). Detailed results per 95 sample are described in supplementary file S4, and detailed results per experiment can be found in supplementary files S5-S31. Multiple aliquots of the same samples were measured. Samples of lava flows were analyzed using step-heating experiments, while for the ash layers usually single or a few grains were fused in one step and analyzed.

The integrated density distribution of these data reveals a peak at around 40.0 Ma. All compiled ages are shown together with the scaled areal extent of mapped units of Sahandi et al. (2014) (see Figure 2C).

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2.2 Compilation of literature data

We combined our newly acquired data with ~370 ages from 60 published studies, including K-Ar, Ar-Ar, U-Pb, Rb-Sr and Re-Os ages (but mainly Ar-Ar and U-Pb; see supplementary file S2). Our age compilation aimed at pre-Quaternary rocks and is incomplete with respect to Quaternary volcanic rocks in Iran. We then used a kernel density plot (Vermeesch, 2012) to integrate all ages from 60-0 Ma, together with our newly acquired data. Ages and their 1σ uncertainties are used as input in the calculation of these distributions. Optimal bandwidth is calculated automatically, and we have set the bin width to 1 Myr. When studies did not report the significance level of their uncertainties, we assumed a 1σ uncertainty. Where possible, Ar-Ar ages were recalibrated to the standard of the Fish Canyon Tuff according to the Kuiper et al. (2008) calibration model. In some cases, original studies did not provide sufficient information for recalibration and then the original ages were used. All details of literature ages and associated references are added in supplementary files S2 and S3.

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The compilation of $^{40}\text{Ar}/^{39}\text{Ar}$ ages from the literature, mostly from extrusive rocks (only 5 Ar-Ar ages are from intrusive rocks), yields a highly similar age density distribution to our dated samples (see Figure 3A), showing a peak at 39.7 Ma. Published U-Pb ages are typically obtained from zircons which provide less accuracy for eruption ages than $^{40}\text{Ar}/^{39}\text{Ar}$ ages from groundmass, plagioclase, sanidine or biotite (Simon et al., 2008), which is reflected in the greater width of the peaks from extrusive U-Pb ages (see Figure 3B). Combined, the Ar-Ar and U-Pb ages obtained from extrusive rocks record a 39.7 Ma peak, along with another sub-peak at 42.8 Ma (see Figure 3C).

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3 Neotethys volcanism and the MECO

Considering that the Neotethys subduction zone has been active since the late Triassic (Arvin et al., 2007), our compilation shows a remarkable clustering of ages during the middle Eocene at ~40 Ma. Estimation of the areal extent of middle Eocene volcanic rocks is done using the shapefiles of Sahandi et al. (2014). For the Eocene, shapefiles are classified as 'Eocene', 'Eocene-Oligocene', 'Late Eocene-Oligocene', 'Middle Eocene', and 'Middle-Late Eocene'. We assumed that shapefiles specified as 'Eocene' had the same proportion of middle Eocene igneous rocks, and thus calculated an areal extent of 38223 km² of middle Eocene igneous rocks.

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Our compilation indicates that many volcanic provinces in Iran were active simultaneously around 40 Ma (see Figure 2C), including the Azerbaijan-Bazman magmatic arc in the west, the Sabzevar zone in northeast Iran (Moghadam et al., 2015) and the Lut block in the east (Pang et al., 2013). Some of the largest volumes of middle Eocene volcanic rocks are located in the Talesh Mountains, where 4 out of 5 exposures with the largest areal extent are mapped (marked in white on Figure 1A). Almost three quarters of U-Pb ages (n=214) in Iran are derived from intrusive rocks (n=148). All ages of the intrusive rocks

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130 together reveal a peak at ~39.8 Ma (Figure 3D), indicating that the peak of middle Eocene extrusive volcanism is also close in time to peak intrusive activity.

It is thus clear that the MECO corresponds to a phase of intense volcanism in the studied area. However, the average error (1σ) of the literature-based ages from 20-60 Ma is 585 kyr, and thus exceeds the duration of the MECO (500 kyr). Furthermore, the exact ages of the peaks in volcanic activity in Figure 2 are sensitive to the number of data points included and are thus not particularly robust – the addition of a few new data points may shift the peaks by thousands of years.

135 4 Volcanic CO₂ emissions in Iran and the MECO

The surface area of Iran covered by middle Eocene volcanic rocks is almost 40,000 km² (Sahandi et al., 2014; Table 2).

~~These volcanic rocks were produced by numerous eruptions throughout the middle Eocene.~~ In the Alborz and Central Iran, middle Eocene volcanic formations are reported to be very thick, with estimates ranging from 3-5 km in the Alborz

Mountains (Stöcklin, 1974), to 6-12 km locally throughout nearly all of Iran (Berberian and King, 1981). More recent estimates of the thickness are 3-9 kilometers (e.g. Morley et al., 2009; Verdel et al., 2011). Extrapolating these thicknesses,

140 this implies a total volume of extrusive middle Eocene volcanics between 1×10^5 and 3.5×10^5 km³ (see Table 2) that potentially produced significant amounts of CO₂.

Our estimates of CO₂ release due to middle Eocene volcanism in Iran are likely underestimates, as there is volcanism in other regions along the Neotethys subduction zone. Unfortunately, the lack of shapefiles of Eocene volcanic and intrusive rocks in Armenia and Azerbaijan, along the Lesser Caucasus Mountains (e.g. Allen and Armstrong, 2008), and plutons and volcanic rocks in Armenia (e.g. Moritz et al., 2016; Sahakyan et al., 2016), hampers calculations on additional CO₂ emissions within these regions.

Due to the absence of quantifications of the relation between the erupted volumes of volcanic rocks and emission of CO₂ in continental arcs, we make a comparison with the Deccan traps, for which this relation has been calculated. The Deccan traps

150 have an estimated eruptive volume of volcanic and volcanoclastic rocks of 1.3×10^6 km³ (Jay and Widdowson, 2008), with an associated emission 4.14×10^{17} mol CO₂ (Tobin et al., 2017). From different estimates of volume and related CO₂ emissions of Tobin et al. (2017), we obtain a linear relation of lava volume (in 10^6 km³)/total CO₂ (in 10^{17} mol) ≈ 0.31 for the Deccan traps.

CO₂ degassing rates for continental arcs may be similar to (Marty and Tolstikhin, 1998), or larger than for continental flood basalts (McKenzie et al., 2016; Wignall et al., 2009). As a conservative starting point, we assume a similar volume versus

155 emission relationship as the Deccan traps, which implies a minimum estimate for CO₂ release from middle Eocene volcanism in Iran between 0.37×10^{17} and 1.10×10^{17} mol (see Table 2), which corresponds to 438-1315 Pg C. Moreover, the amount of CO₂ released during volcanic episodes has been shown to increase substantially if eruptions occur among carbonate-rich sediments (Lee et al., 2013; Lee and Lackey, 2015). For example, CO₂ released from carbonate sediments during the emplacement of the Emeishan large igneous province in the end-Guadalupian was estimated to be 3.6-8.6 times

160 higher than the amount of CO₂ released by volcanic outgassing alone (Ganino and Arndt, 2009). Indeed, the Eocene extrusive volcanism in Iran erupted through significant amounts of carbonate-rich rocks of Jurassic, Cretaceous, and

Deleted: extrusive

Deleted: extrusive

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Deleted: Due to the limited number of studies quantifying the relation between deposited volume of volcanic rocks and the emission of CO₂, we make a comparison with the Deccan traps, for which this relation has been calculated

Paleogene age (e.g. Berberian and King, 1981). As a result, carbon release associated with the production of volcanic rocks in Iran could be much larger, potentially ranging from 1578 to 11,308 Pg C (see Table 2). This range of CO₂ emissions is compatible with the carbon cycle imbalance that drives the MECO in simple carbon cycle simulations constrained by available proxy data (roughly 2000-4000 Pg C; Henehan et al., 2020; Sluijs et al., 2013; van der Ploeg et al., 2018). Erosion has affected the entire Iranian plateau, and could have eroded away significant volumes of Eocene volcanic rocks. Morley et al. (2009) and Ballato et al. (2011) note that clasts in the Lower and Upper Red formation (Oligocene-Miocene age), which in many places overlie Eocene volcanics, are for a large part made up of eroded Eocene volcanic rocks. Original thicknesses of Eocene volcanic rocks in Iran could thus have been larger, making our CO₂ output estimate a minimum estimate.

180 **5 Future perspectives**

There are several obstacles in solidifying the link between warming during the MECO and volcanism in the Neotethys subduction zone. First of all, continental arcs are generally active for (tens of) millions of years, while the MECO has a duration of 500 kyr. Moreover, this duration is shorter than common uncertainties for radiometric ages in the Eocene, complicating the establishment of a causal relationship. This is important because a driver for the MECO requires excess CO₂ input only during the ~500 kyr spanning the MECO, and not during the time surrounding it (Sluijs et al., 2013). This is also supported by the drop in global ocean osmium isotope ratios, which is specifically associated with the MECO interval (van der Ploeg et al., 2018). Secondly, Iran is a relatively understudied area compared to other (continental) arcs. As a result of this, the amount of radiometric ages is low, with on average about 1 radiometric age for every several hundred km² of outcrop.

190 Therefore, the relation in time between the MECO and Neotethys arc flare-up calls for the development of much better age constraints of the volcanic deposits in Iran and this is certainly feasible. While most flare-ups have to be studied via their intrusive roots, as the extrusive record is removed through erosion (Ducea and Barton, 2007; de Silva et al., 2015), the extrusive record in Iran is extensive so that the ages can be mapped in high detail. Moreover, the respective roles of intrusive and extrusive rocks can be assessed to estimate the amount of volatiles of the igneous rocks, and sedimentological studies can provide minimum estimates on how much extrusive rock has been lost through erosion. This would help solve the question if CO₂ input rates across from the Neotethys flare-up were truly excessive and caused a net addition of CO₂ during the MECO.

200 **6 Conclusions**

We provide new Ar-Ar ages from volcanic rocks of the Azerbaijan-Bazman Arc in Iran and combine these with literature data to show that a flare-up of continental arc volcanism in Iran peaked about 40 Ma ago, conspicuously close to the Middle Eocene Climatic Optimum. We estimated volumes of middle Eocene volcanism in Iran to be between $1 \cdot 10^5$ and $3.5 \cdot 10^5$ km³. We compared the volume of middle Eocene volcanics in Iran to that of the Deccan traps and estimate that between 438

and 1315 Pg of carbon in the shape of CO₂ was released during deposition. Taking into account the fact that all volcanism
205 occurred in shallow marine basins and erupted in and through pre-existing carbonate-rich rocks, CO₂ release might have
been between 1578 and 11308 Pg. Although the flare-up must be dated much better to establish its chronological relation
with the MECO in more detail, we consider it a plausible major contributor to greenhouse warming during the MECO.

7 Supplementary materials

210 Examples of scans of thin sections are supplied in supplementary file S1. All details of literature ages and associated
references are added in supplementary files S2 and S3. A detailed description of Ar-Ar results per sample is provided in
supplementary file S4. Supplementary files S5-S31 show the results of the ⁴⁰Ar/³⁹Ar geochronology per experiment. S32
shows an extended version of the literature age plot of Figure 2C.

215 **Author contributions:** Fieldwork was undertaken by AvdB, MH and WK. AvdB, KFK and MH performed Ar-Ar dating.
Data analysis was performed by AvdB, KFK, RvdP, MJC and AS. All authors contributed to scientific discussions and were
involved in writing the manuscript.

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