Reviewer #1

Dear van der Boon,

Thanks for the clarification. I agree with most of your points. Here I have two more suggestions that may help you to strengthen your points.

1. Since you have collected many age data with GPS information and highlighted your new data in figure 1b, I suggest that you plot all available mid-Eocene age on your figure 1a. The time-space distribution of the mid-Eocene rock in Iran would help readers to understand how widespread the Eocene rocks.

This is a good suggestion and we have attempted to do this. However, we found that the more than 420 radiometric ages from 72 studies clutter up the figure so much that it becomes unreadable, and/or font too small to read. To accommodate for the suggestion of the reviewer, we have added a file to the supplementary information that contains all the ages of the supplementary information with their GPS locations. This file is our new supplementary file S4, and we have added to the text in section 7 "S4 is a .kmz file that contains the GPS locations of the literature ages (except of Shafaii Moghadam et al., (2020), who did not provide GPS locations), and can be opened in Google Earth."

2. New age clusters at 40 Ma is in indeed close to the MECO. However, the age cluster does not guarantee the large volume of the volcanic eruption. If a net input of CO2 causes the MECO, you would expect increasing volcanism from Paleocene (or early Eocene) to 40 Ma. Since you have the shapefile information, I suggest you make a plot: Area of volcanism vs. age. If the volume of the middle Eocene gets to the maximum, which would make your argument more convincing.

We do not fully understand this comment. A pulse increase in volcanism at ~40 Ma is required to explain the MECO warming. In addition, it seems to us that the plot suggested by the reviewer is our Figure 2C.

Reviewer #3

The manuscript "Exploring a link between the Middle Eocene Climatic Optimum and Neotethys continental arc flare-up" suggests the existence of a possible causal relationship between the Eocene arc volcanism associated with the subduction of the Neotethys ocean and the Middle Eocene Climatic Optimum (MECO). To test this hypothesis the authors provides new Ar-Ar ages of Middle Eocene volcanics exposed in NW Iran, compiles available Ar-Ar and Zircon U-Pb ages of Cenozoic intrusive and effusive rocks exposed all around Iran, and estimate the volume of CO2 that might have been released in the atmosphere during the flare up. This working hypothesis represents an intriguing idea, however, it has not been clearly demonstrated in this version of the manuscript.

My main point is the same raised by Reviewer 1 "the successful link between the arc volcanism and climate change depends on how much carbon dioxide has been outputted" around the MECO. Furthermore, Reviewer 1 wrote: "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." This is a key point that the authors did not address.

Our main reply to this reviewer's comment is that we cannot prove, nor do we intend to, that the volcanism in Iran ***caused*** *the MECO. We have phrased carefully to convey that point, both in the title* *and throughout the manuscript. We explore whether this volcanism could have had an impact on global climate in the middle Eocene, since there has been a decade long search for volcanism that coincided with the MECO. We are open and honest about the limitations of this study, and present those clearly in the manuscript. Although the constraints that currently exist on both the timing and the areal extent of Eocene volcanism in Iran prevent a definitive conclusion on a causal relationship, they are good enough to make the 'back of the envelope' calculations that we present in this manuscript, but, again, the uncertainties are large and, in our view, clearly described. In the very least, we feel that the data compilation that we present here is large enough to support our assessment of the Eocene flare-up in Iran to broadly correspond to the MECO.*

The authors replied that: "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." I have not found in the original paper any analytical data supporting this conclusion. Furthermore the authors replied that: "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" This new text does not indicate the thickness of volcanics and volcaniclastics deposited around the MECO (or let's say at 40±2 Ma).

Unfortunately, the amount of radiometric ages obtained on volcanic rocks in Iran is currently not large enough to assess exactly which part of the volcanic succession erupted within 2 million years around 40 Ma. This remains one of the uncertainties, as described in section 5. With regards to the thickness of the Eocene volcanic rocks, much of the mapping has been done in the 1960's, and many of the reports are not available online, or even in English. However, there are more assessments of thickness based on geological maps and cross-sections, and we have added a reference to Iwao and Hushmand-Zadeh (1971)*, who show a generalised lithostratigraphic column of the Karaj Formation in the Alborz Mountains. This column shows that the Karaj Fm has a thickness of around 10 kilometres. We have added to the text the following paragraph and references (lines 149-158): "These estimates are supported by geologic maps and their descriptions that are based on extensive fieldwork. Estimates from maps range mostly between 2 and 7 kilometres. On the lower side are Saein Qaleh (Kholghi Khasraghi, 1994), Saveh (Ghalamghash et al., 1998b), Kuhpayeh (Radfar et al., 2002), with thicknesses of ~2 kilometres, Tafresh (Hadjian et al., 1999) with ~3 kilometres, then Meyamey (Amini Chehragh and Ghalamghash, n.d.), Tarom (Hirayama et al., 1966) and Kalateh (Jafarian, n.d.) with around ~4 kilometres, while Kajan (Amini and Amini Chehragh, 2001), Kahak (Ghalamghash et al., 1998a) and Lahrud (Babakhani et al., 1991) mention thicknesses of the Eocene volcanic succession of around ~6 kilometres, and Bardsir (Mohajjel Kafshdouz and Khodabandeh, 1992) of around 7 kilometres. On the other hand, Iwao and Hushmand-Zadeh, (1971) show a generalised lithostratigraphic log of the Karaj formation, and mention that the succession reaches a thickness of more than 10 kilometres in the Alborz Mountains." We have adjusted our Table 2 to include more estimates of thicknesses, and we calculate CO_2 estimates for the entire range of thicknesses between 2 and 10 kilometres. We have added to the text (lines 158-160) "In Table 2, we calculate how much CO_2 could have been released through formation of different volumes of volcanic rocks. We calculate this for a range of thicknesses between 2 and 10 kilometres." and (lines 188-192) "Table 2 shows that middle Eocene volcanic rocks with thicknesses between 2 and 7 kilometres, and a contribution from limestones, give estimates that lie within the range expected for the MECO (marked in green in Table 2). There could have been a contribution to CO_2 through skarn formation by intrusive activity, which clusters around 40.5 Ma (see Figure 3D), although we currently lack the constraints to quantitatively assess this." and (lines 188-190): "Table 2 shows that middle Eocene* *volcanic rocks with thicknesses between 2 and 7 kilometres, and a contribution from limestones, give estimates that lie within the range expected for the MECO (marked in green in Table 2).”.*

In section 4 the authors calculate the volume of volcanic rocks that have been erupted during the flare up (3-9 km of thickness times 40.000 km2) and based on a “linear relationship of a lava volume” (see line 150) they estimate the amount of released CO2. It is not clear to me what is the age of the Middle age volcanics considered in the calculation. From figure 2 and from the text I have the impression that the authors considered all the magmatic rocks produced during the entire flare up, which lasted about 20 million years (from ca. 55 to 35 Ma), and not the thickness of the 40±2-My-old magmatic rocks. In the text, the authors recall also table 2, but I could not find it in the text. As specified above, the amount of magmatic rocks emplaced around the MECO is a crucial point. Without an idea of such a thickness, the released CO2 cannot be estimated, and the working hypothesis cannot be tested.

We explained in lines 118-122 that we calculate the amount of middle Eocene volcanic rocks in the following way: “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 km^2 of middle Eocene igneous rocks.” So we calculate estimated volumes for the middle Eocene (10 Myr), as this is the maximum resolution that we can get from the shapefiles. With only one radiometric age for every few hundred square kilometres in Iran, this is currently as good as it will get without a stellar amount of new radiometric ages. We have now rephrased this, in the hope that it is clearer. It now reads (lines124-130): “According to the geologic maps, 54% of all area covered by volcanic rocks in Iran is of Eocene age. For the Eocene, shapefiles are classified as ‘Eocene’, ‘Eocene-Oligocene’, ‘Late Eocene-Oligocene’, ‘Middle Eocene’, and ‘Middle-Late Eocene’. More than half is marked as ‘Eocene’ and not specified further, but of the rest that is specified, almost half is ‘Middle Eocene’. Assuming that the unspecified Eocene rocks have approximately the same age distribution as the specified Eocene rocks, we estimate that roughly half of the Eocene volcanic rocks in Iran and a quarter of the total area covered by volcanic rocks in Iran is of middle Eocene age. We use these areas to estimate the volumes of volcanic rocks formed in the middle Eocene.”

Curiously, if the authors look at the Zachos et al., curve (δ18O curve vs age), they will see that the flare up in Iran coincides with the progressive Eocene cooling that culminates with the sharp temperature drop at the Eocene-Oligocene boundary (actually I think that such a curve, which is the base of all paleoclimatic reconstructions, should be shown also in this manuscript). To me this lack of correlation suggests that, although voluminous, the entire magmatic flare up in Iran did not have a strong impact on global climate, or at least that did not produce a change in the long-term global cooling trend.

As our radiometric age compilation shows in Figure 2C, the amount of radiometric ages drops rapidly between 35 and 32 Ma (the slope is nearly flat here). Also Figure 3 shows a drastic drop in igneous activity around the Eocene-Oligocene transition. The flare-up in Iran precedes the Eocene-Oligocene transition by millions of years. On a side note, several of the authors are marine stratigraphers and paleoceanographers with ample experience on both the MECO and the Eocene-Oligocene Transition.

If the authors want to demonstrate a causal relationship between arc volcanism and MECO they need to document an increase in magmatic flux at 40±2 Ma. So far, nobody as really demonstrated it. To me, at least as first approach, they should look at few stratigraphic sections all around the country, measure their thickness, extract the depositional ages and then estimate changes in magmatic flux through time. I think that is the most direct way to test such a working hypothesis. After that, they may look at the areal distribution on Middle Eocene rocks, assuming that these are really Middle Eocene rocks that were deposited around the MECO and not during the entire flare up.

We fully agree. We have incorporated this point in section 5 (lines 214-215): "Acquisition of radiometric ages throughout sections that cover the entire Eocene volcanic succession could aid in quantification of magmatic flux over time."

Indeed, the Peshtasar Formation is a good target because available ages from Vincent et al (2005), recalibrated by the authors, indicate deposition of an up to 1.4-km-thick sequence of volcanics and volcaniclastics between 41 and 39 Ma. Note that we are talking about 1.4 km of volcanics and volcaniclastics and not 3 to 9 km. Similar work should be done in other areas.

The Peshtasar formation is certainly important but, importantly, it is only ***part of*** *the entire (middle?) Eocene succession in the Talysh Mountains. The Peshtasar Formation is more than 2 kilometres thick* (van der Boon et al., 2017) *and consists only of basalts and sills, likely formed in three very short pulses of intense volcanism. There is another formation underneath this, the Kosmalyan formation, which also consists of another large amount of volcanic and volcaniclastic rocks. Although this formation has not been dated, nor studied in detail, Vincent et al. (2005) estimate the thickness of the Kosmalyan formation as more than 7 kilometres, with an estimated age of late early to early middle Eocene. This could thus mean that the middle Eocene succession in the Talysh of Azerbaijan is around 9 kilometres thick, which would be in line with findings from the Talesh and Alborz of Iran. Collectively, we passionately agree with the reviewer that the entire Eocene succession in Iran warrants detailed study and hope that our paper will spur enthusiasm of the geology community to study Eocene volcanic rocks in Iran.*

I understand that the compilation provided by the authors try to overcome the paucity of stratigraphic information available in literature, but that strategy is biased toward the sampled stratigraphic intervals. It may be correct for intrusive rocks because there might be a cluster of ages (assuming that the cooling recorded by the Ar-Ar system occurred within 1-2 Million years) indicating a specific episode of magma emplacement and possibly an overproduction of magma. However, according to available data (Verdel et al., 2011, is probably the best reference) effusive and pyroclastic rocks could have been deposited between 55 and 35 Ma at rather uniform rates . Except the work of Vincent et al., 2005, there are no studies pointing toward an increase in the magmatic flux around the MECO.

We agree with the reviewer that our compilation is likely biased towards certain accessible areas and intervals, and we have added to the text the following paragraph (lines 196-200): "Despite the fact that sampling biases (i.e. sampling is often focused on easily accessible sites and certain time periods) can never be avoided, our compilation of radiometric ages shows a good correlation to the geologic maps, in the sense that the radiometric ages confirm that the flare-up took place during the middle Eocene. We note that the Miocene peak (Figure 3C) is relatively high compared to the Eocene, which could be caused by a sampling bias, as the geologic maps (Sahandi et al., 2014) indicate that only 2-4% of Iran is covered by Miocene volcanic rocks." We disagree with the reviewer that there are no studies besides Vincent et al. (2005) that point towards an increase in magmatic flux around the MECO. On the contrary, field studies have often suggested that the middle Eocene part makes up the bulk of the Eocene succession (e.g. Glaus, 1965). *Also* Davoudzadeh et al. (1997) *mention "Extensive volcanism with a wide range of composition started in Upper Cretaceous and continued throughout the Cainozoic with the climax in Middle Eocene time." In our previous response to reviewer 1, we also mentioned that 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." Furthermore,* Verdel (2009) *shows in Figure 5 of Chapter 3 that most of the flare-up is during the middle Eocene for North, West and East Iran, and only in Central Iran extends also into the lower Eocene. Additionally, a middle Eocene increase in volcanism can be seen from the slope of the radiometric ages in our plot in Figure 2C, which is much steeper during the Lutetian and Bartonian, indicating that there are many more radiometric ages in this time period than in the times before and after the middle Eocene. We have added to the introduction (lines 58-60): "Field studies have often suggested that the middle Eocene part makes up the bulk of the Eocene succession (e.g. Glaus, 1965), and volcanism climaxes during middle Eocene time (Berberian and King, 1981; Davoudzadeh et al., 1997; Verdel, 2009)."*

Finally, concerning the compilation and the genesis of magma (see comment 1 of Reviewer 2), I suggest looking at a recent publication of Rabiee et al., in Gondwana Research titled: "Long-lived, Eocene-Miocene stationary magmatism in NW Iran along a transform plate boundary"; there, new ages of intrusives and a similar compilation is provided.

We thank the reviewer for this suggestion. Consequently, we have added the U-Pb ages of this and of another 11 studies (Almasi et al. (2019), Etemadi et al. (2020), Javidi Moghadam et al. (2019, 2020), Khaksar et al. (2020), Maleki et al. (2019), Mazhari et al. (2020), Rabiee et al. (2019, 2020), Sepidbar et al. (2019), Shafaii Moghadam et al. (2020) and Simmonds (2019)) to our compilation, which now consists of 72 papers and more than 420 ages. We updated our Supplementary files S2, S3 and S33 accordingly. We made new versions of figures 2C and figures 3B, 3C and 3D. We have adapted ages for the peaks in the text accordingly.

Here are few minor points:
1) In the abstract, the authors suggest that magma emplacement in carbonaceous rocks may have increased the total amount of CO2 released. This is not really addressed in the text except in lines 159-161 where the authors write: "Indeed, the Eocene extrusive volcanism in Iran erupted through significant amounts of carbonate-rich rocks of Jurassic, Cretaceous, Paleogene age (e.g. Berberian and King, 1981)". By looking at geologic maps in NW Iran, it seems to me that most of Eocene intrusions are found within Eocene volcanics and volcaniclastics (meaning that they intruded at shallow depth) rather than in Paleo-Mesozoic carbonates (while Paleogene carbonates are rather thin). I cannot see evidences of intrusions in carbonaceous lithology based on available geologic maps. It may be true, with erosion that has not brought yet these rocks at the surface, but currently there is not any evidence for that.

We do not really understand this comment. The reviewer refers to our lines 159-161, where we talk about extrusive rocks, but then continues to state that Eocene intrusions are found within Eocene volcanics and volcaniclastics. We agree with the reviewer and there is no contrast here. We have however removed the pleonasm 'extrusive volcanism' throughout the manuscript.

We have, however, added some more references and a paragraph on the association of the volcanic rocks with shallow marine carbonate-rich rocks, which now reads (lines 179-184): "Indeed, the Eocene volcanism in Iran erupted in shallow marine basins, and through significant amounts of carbonate-rich rocks of Jurassic, Cretaceous, and Paleogene age (e.g. Berberian and King, 1981). Glaus, (1965) mentions that middle Eocene limestones occur as lenticular masses within the basaltic flows, or as consistent horizons associated with tuffs. Verdel (2009) shows that Eocene volcanic rocks are formed in close association with Eocene limestones in north, west and east Iran. This is also the case in central Iran, which can be seen from geologic maps, such as the one from Qom (Emami, 1981)."

2) Reviewer 1 suggested also to look also at other regions as possible sources of arc volcanism around the MECO. Of course, the first region to look at would be the entire Middle East, which represents the upper plate of the Neothetys subduction system (Turkey, Armenia, Georgia, Azerbaijan). The authors replied ”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”
I do not think that the lack of shape files hampers the calculations. The lack of a geologic map with clear ages of volcanics hampers the calculations. If you have a geologic map with good age information you can easily digitize the contours of the Middle Eocene volcanics and create a shape file.

These are all good suggestions and this would be good follow-up study. Unfortunately, we do not have such maps. The maps we have for Azerbaijan and Armenia are very low resolution (and we only have one of each country), and stand in stark contrast to the more than 500 maps we have from Iran.

Note, however, that there is still a large volume of volcanic and volcaniclastic rocks buried below late Cenozoic sediments that are difficult to estimate. This is particularly true in Central Iran where depositional processes are dominant and localized exhumation hasn’t yet exposed the Eocene volcanics. This means that any estimates based on outcrops will be always a very minimum value.

We agree with the reviewer, and mention this in lines 174-178 (new lines 192-196): “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 CO_2 output estimate a minimum estimate”.

Finally, I am not a geochemist, so I cannot comment on the calculations for estimating the released CO2, but I guess that the starting point is a reliable estimate of the volume of volcanics and volcaniclastics ejected around the MECO.

I hope these comments will help. Good luck.

We thank the reviewer for their comments that have prompted us to rethink a number of issues, further complete our database and improve our manuscript.

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

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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 igneous rocks in all volcanic provinces of 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 CO_2 that could have been released at this time corresponds to between 1052 and 12,565 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.

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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 CO_2 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 $\delta^{13}C$ 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 (e.g. 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). Field studies have often suggested that the middle Eocene part makes up the bulk of the Eocene succession (e.g. Glaus, 1965), and volcanism climaxes during middle Eocene time (Berberian and King, 1981; Davoudzadeh et al., 1997; Verdel, 2009).

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

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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 continental arc flare-up.

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

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

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 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 $^{40}Ar/^{39}Ar$ 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 or glass and plagioclase or biotite could be separated.

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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 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 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 $^{40}Ar/^{39}Ar$ 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 sample are described in supplementary file S5, and detailed results per experiment can be found in supplementary files S6-S32. Multiple aliquots of the same samples were measured. The integrated density distribution of these data reveals a peak at around 40.0 Ma (Figure 2B). 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 more than 420 ages from 72 published studies, including K-Ar, Ar-Ar, U-Pb, Rb-Sr and Re-Os ages (but mainly Ar-Ar and U-Pb; see supplementary files S2 and S3). Our age compilation aimed at pre-Quaternary rocks and is incomplete with respect to Quaternary and pre-Paleogene igneous 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 $^{40}Ar/^{39}Ar$ 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 $^{40}Ar/^{39}Ar$ 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 wide peak around 42 Ma, with two sub-peaks at 43.4 and 39.4 Ma. Two smaller peaks at 29.8 and 17.1 Ma are apparent (see Figure 3C). Intrusive activity also peaks around the same time, with radiometric ages from intrusive rocks (n=201) showing a peak at 40.5 Ma, with another sub-peak at 36.6 Ma (Figure 3D). Smaller peaks in intrusive activity are present at 29.7 and 19.9 Ma.

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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), who made a compilation of geologic maps. According to the geologic maps, 54% of all area covered by volcanic rocks in Iran is of Eocene age. For the Eocene, shapefiles are classified as ‘Eocene’, ‘Eocene-Oligocene’, ‘Late Eocene-Oligocene’, ‘Middle Eocene’, and ‘Middle-Late Eocene’. More than half is marked as ‘Eocene’ and not specified further, but of the rest that is specified, almost half is ‘Middle Eocene’.

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Assuming that the unspecified Eocene rocks have approximately the same age distribution as the specified Eocene rocks, we estimate that roughly half of the Eocene volcanic rocks in Iran and a quarter of the total area covered by volcanic rocks in Iran is of middle Eocene age. We use these areas to estimate the volumes of volcanic rocks formed in the middle Eocene. We thus assumed that shapefiles specified as 'Eocene' had the same proportion of middle Eocene igneous rocks, and calculated an areal extent of 38223 km^2 of middle Eocene igneous rocks.

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 (Shafaii 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_{total}=329) in Iran are derived from intrusive rocks ($n_{intrusive}$=239). All ages of the intrusive rocks together reveal a peak at ~40.5 Ma (Figure 3D), indicating that the peak of middle Eocene 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.

4 Volcanic CO_2 emissions in Iran and the MECO

The surface area of Iran covered by middle Eocene volcanic rocks is almost 40.000 km^2 (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). These estimates are supported by geologic maps and their descriptions that are based on extensive fieldwork. Estimates from maps range mostly between 2 and 7 kilometres. On the lower side are for example Saein Qaleh (Kholghi Khasraghi, 1994), Saveh (Ghalamghash et al., 1998a), and Kuhpayeh (Radfar et al., 2002), with thicknesses of ~2 kilometres, Tafresh (Hadjian et al., 1999) with ~3 kilometres, then Meyamey (Amini Chehragh and Ghalamghash, n.d.), Tarom (Hirayama et al., 1966) and Kalateh (Jafarian, n.d.) with around ~4 kilometres, while Kajan (Amini and Amini Chehragh, 2001), Kahak (Ghalamghash et al., 1998b) and Lahrud (Babakhani et al., 1991) mention thicknesses of the Eocene volcanic succession of approximately 6 kilometres, and Bardsir (Mohajjel Kafshdouz and Khodabandeh, 1992) of around 7 kilometres. On the other hand, Iwao and Hushmand-Zadeh, (1971) show a generalised lithostratigraphic log of the Karaj formation, and mention that the succession reaches a thickness of more than 10 kilometres in the Alborz Mountains. In Table 2, we calculate how much CO_2 could have been released through formation of different volumes of volcanic rocks. We calculate this for a range of thicknesses between 2 and 10 kilometres. Extrapolating these thicknesses, this implies a total volume of middle Eocene volcanic rocks between

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7.6*10^4 and 3.8*10^5 km^3 (see Table 2) that potentially produced significant amounts of CO_2. Our estimates of CO_2 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 CO_2 emissions within these regions.

Due to the absence of quantifications of the relation between the erupted volumes of volcanic rocks and emission of CO_2 in continental arcs, we make a comparison with the Deccan traps, for which this relation has been calculated. The Deccan traps have an estimated eruptive volume of volcanic and volcaniclastic rocks of 1.3*10^6 km^3 (Jay and Widdowson, 2008), with an associated emission 4.14*10^{17} mol CO_2 (Tobin et al., 2017). From different estimates of volume and related CO_2 emissions of Tobin et al. (2017), we obtain a linear relation of lava volume (in 10^6 km^3)/total CO_2 (in 10^{17} mol) ≈ 0.31 for the Deccan traps.

CO_2 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 emission relationship as the Deccan traps, which implies a minimum estimate for CO_2 release from middle Eocene volcanism in Iran between 2.34*10^{16} and 1.22*10^{17} mol (see Table 2), which corresponds to 292-1461 Pg C. Moreover, the amount of CO_2 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, CO_2 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 higher than the amount of CO_2 released by volcanic outgassing alone (Ganino and Arndt, 2009). Indeed, the Eocene volcanism in Iran erupted in shallow marine basins, and through significant amounts of carbonate-rich rocks of Jurassic, Cretaceous, and Paleogene age (e.g. Berberian and King, 1981). Glaus, (1965) mentions that middle Eocene limestones occur as lenticular masses within the basaltic flows, or as consistent horizons associated with tuffs. Verdel (2009) shows that Eocene volcanic rocks are formed in close association with Eocene limestones in north, west and east Iran. This is also the case in central Iran, which can be seen from geologic maps, such as the one from Qom (Emami, 1981). As a result, carbon release associated with the production of volcanic rocks in Iran could be much larger, potentially ranging from 1052 to 12,565 Pg C (see Table 2). This range of CO_2 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). Table 2 shows that middle Eocene volcanic rocks with thicknesses between 2 and 7 kilometres, and a contribution from limestones, give estimates that lie within the range expected for the MECO (marked in green in Table 2). There could have been a contribution to CO_2 through skarn formation by intrusive activity, which clusters around 40.5 Ma (see Figure 3D), although we currently lack the constraints to quantitatively assess this. 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.

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Original thicknesses of Eocene volcanic rocks in Iran could thus have been larger, making our CO_2 output estimate a minimum estimate. Despite the fact that sampling biases (i.e. sampling is often focused on easily accessible sites and certain time periods) can never be avoided, our compilation of radiometric ages shows a good correlation to the geologic maps, in the sense that the radiometric ages confirm that the flare-up took place during the middle Eocene. We note that the Miocene peak (Figure 3C) is relatively high compared to the Eocene, which could be caused by a sampling bias, as the geologic maps (Sahandi et al., 2014) indicate that only 2-4% of Iran is covered by Miocene volcanic rocks.

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 CO_2 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 km^2 of outcrop.

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. Acquisition of radiometric ages throughout sections that cover the entire Eocene volcanic succession could aid in quantification of magmatic flux over time. 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 constrain CO_2 input rates across from the Neotethys flare-up to a narrower interval around the MECO.

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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 $7.6*10^4$ and $3.8*10^5$ km^3. We compared the volume of middle Eocene volcanics in Iran to that of the Deccan traps and estimate that between 292 and 1461 Pg of carbon in the shape of CO_2 was released during deposition. Taking into account the fact that all volcanism occurred in shallow marine basins and erupted in and through pre-existing carbonate-rich rocks, CO_2 release might have

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been between 1052 and 12,565 Pg. Thicknesses of the middle Eocene volcanic succession between 2 and 7 kilometres, with a contribution from carbonate-rich rocks result in estimates of released carbon that are in line with estimates for the MECO. 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.

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7 Supplementary materials

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. S4 is a .kmz file that contains the GPS locations of the literature ages (except of Shafaii Moghadam et al., (2020), who did not provide GPS locations), and can be opened in Google Earth. A detailed description of Ar-Ar results per sample is provided in supplementary file S5. Supplementary files S6-S32 show the results of the $^{40}Ar/^{39}Ar$ geochronology per experiment. S33 shows an extended version of the literature age plot of Figure 2C.

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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. We thank two anonymous reviewers for their comments on an earlier version of this manuscript, and three anonymous reviewers for their comments that have improved this manuscript.

Competing interests: The authors declare no competing interests.

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