Appendix C. Geochemical considerations regarding the assumed abundances of U, Th, and K in the mantle

C1. Geochemical Introduction

Geodynamic processes are intrinsically tied to geochemical realities, meaning that the accuracy of numerical modelling of geodynamic processes is dependent on the quality of the geochemical assumptions that underlie the modelling. A pivotal question is the *initial* chemical composition of the mantle, where apart from primordial heat, the distribution of the abundances of U, Th, and K, the three significant heat-producing elements within the mantle, is crucial for powering the mantle motor.

Palme and O'Neill (2003) reviewed the distribution of major elements in the mantle and compared the Al/Si, Mg/Si, Fe/Si, Na/Si, Zn/Si, S/Si, and O/Si ratios of the Sun and the major group of chondritic meteorites, namely CI, CM, CO, CV, H, L, LL, EH, and EL. This research highlighted a good match between solar and CI meteorite compositions, albeit with certain deviations; for example, both the Earth and carbonaceous chondrites are depleted in manganese. This suggests that Earth and carbonaceous chondrites both record fractionation processes that occurred within the inner solar system.

C2. Should we use a non-chondritic Earth model?

Boyet and Carlson (2005, 2006) analysed measurements of ^{142}Nd/^{144}Nd ratios of kimberlites, carbonatites, komatiites, ocean island basalts, and basalts from the Pacific and Indian oceans. The terrestrial Nd standard has a ^{142}Nd/^{144}Nd value that is 20 ppm or 18±5 ppm (Jackson and Carlson, 2012) higher than that in chondrites. In addition, terrestrial rocks younger than 3.5 Ga have virtually identical ^{142}Nd/^{144}Nd ratios. This suggests three possibilities:

I. The Earth accreted from non-chondritic material, or

II. The Earth accreted from chondritic meteorites or from differentiated planetesimals that previously formed from chondritic meteorites. The low ^{182}W/^{184}W ratios of iron meteorites (Harper and Jacobsen, 1996; Kleine et al., 2002; Schoenberg et al., 2002) indicate that differentiated planetesimals contributed a significant amount of material to the early Earth, in turn suggesting that this early Earth underwent very early differentiation. This most likely formed a large early-depleted reservoir (EDR) along with a complementary smaller early-enriched reservoir (EER), thereby explaining the ^{142}Nd/^{144}Nd observations outlined above.

There are two sub-cases, as follows. IIa. The EER was near the Earth's surface and was lost very early through collisions (O'Neill and Palme, 2008). IIb. Some unknown mechanism kept the EER in the deeper mantle, which prevents us from obtaining samples.

Cases I and IIa have similar consequences for the thermal and chemical evolution of the Earth. Boyet and Carlson (2006) emphasised that the abundances of U, Th, and K in the EDR represent only 60% of the abundances of these elements in a chondritic mantle. This means that a high percentage of the laterally averaged surface heat flow, (qob), would be derived from primordial heat. This in turn suggests that present-day laterally averaged heat flow (q_c) at the core–mantle boundary (CMB) would be much higher than previously thought. For case IIa, O'Neill and Palme (2008) reported that satisfying Sm–Nd and Lu–Hf isotopic constraints indicates that U and Th abundances in the Earth's mantle are some 10%–30% lower than within chondritic mantle. White and Morgan (2011) also indicated that by meeting these requirements, steady-state geodynamics or geodynamics with *small* variations in the spatially averaged kinetic energy of mantle creep are impossible. Cases I and IIa, and any case involving *heavy* depletion, mean that the present-day rate of slab subduction is not sustainable, or, in the case of *moderate* depletion, the present-day mantle is in a phase of faster than normal plate motion. It is also possible that the EDR has been identified, as the ϵ^{143}Nd value of high-^{3}He/^{4}He Baffin Island lavas (Stuart et al., 2003; Starkey et al., 2009) is similar to the ϵ^{143}Nd value of the EDR (Jackson et al., 2010; Jackson and Carlson, 2012).

It is feasible to run the Terra code with a chemical differentiation extension to account for these different initial assumptions regarding geochemistry. Calcium–aluminium rich inclusions (CAIs) within carbonaceous chondritic meteorites show only small variations in age determinations, with Jacobsen et al. (2008) obtaining an age of 4567.6 ± 0.4 Ma for a CAI from the Allende chondrite. Here, we follow Carlson and Boyet (2009) and assume an age of 4567.6 Ma for the start of the radioactive decay of U, Th and K in our computations. The ^{182}W/^{184}W ratio of the majority of iron meteorites is almost identical to the initial ^{182}W/^{184}W of CAI, meaning that Carlson and Boyet (2009) assumed that the metal–silicate differentiation occurred within less than 1 Ma of formation of the meteorites. According to Wood et al. (2006), the "depleted" ^{182}W isotopic composition of iron meteorites also indicates that core formation within asteroidal parent bodies occurred before 5 Ma had lapsed since formation of the bodies. However, the accretion of the Earth from planetesimals with metallic iron cores and segregation of the Earth's core took 30–40 Ma. We do not use these exact numbers in our dynamic model because we can only start a run when the mantle is almost entirely crystalline. This results from the fact that our determination of the viscosity profile is based on solid-state considerations and that the mantle has been essentially solid throughout the majority of Earth's history. Running our model with a mantle containing very large volumes of fluid results in a numerical breakdown.

C3. Recent evidence of a chondritic mantle

In addition to the manganese depletion mentioned above, the composition of the terrestrial planets deviates from the mean composition of the solar system in a number of other ways. Marty et al. (2010, 2011) reported that the terrestrial values of $^{15}N/^{14}N = 3.676 \times 10^{-3}$ and $D/H = (1.5 \pm 0.3) \times 10^{-4}$ are similar to the $^{15}N/^{14}N$ and D/H values of Venus, the interior of Mars, and to CI and CM carbonaceous chondrites. We assume that CI and CM chondrites are the most primitive of the carbonaceous chondrites. Furthermore the $\Delta^{17}O$ of the Earth is close to that of CI carbonaceous chondrites. The $^{15}N/^{14}N$ ratio of the solar wind is $(2.178 \pm 0.024) \times 10^{-3}$ (Marty et al., 2011), close to the $^{15}N/^{14}N$ ratio of Jupiter. This means that the Earth, and CI and CM chondrites are all enriched in ^{15}N relative to the protosolar nebula (PSN) as long as the composition of the present-day Earth is considered to be largely consistent with the composition of this nebula. The D/H value of the PSN is $(2.5 \pm 1.5) \times 10^{-5}$, meaning that both the Earth and the CI and CM chondrites are also enriched in deuterium relative to the PSN. Marty (2012) documented that water, carbon, neon, argon, and krypton are in *chondritic* relative proportions in the bulk Earth. Formerly, it was suggested that these and other volatiles in the Earth are derived from comets. However, Marty et al. (2010) indicated that comets have $^{15}N/^{14}N$ ratios of 7.5×10^{-3} and D/H ratios of 3.0×10^{-4}, suggesting that the Earth is essentially derived from carbonaceous chondrites or asteroidal parents derived from chondritic matter, presumably mixed with a small solar component, as accretion took place earlier than was previously thought.

Combining the clustering of zircon ages with the results of this study indicates that continental crustal growth was episodic, including during the Archean. Each episode of continental growth starts with rocks having a *chon-*

dritic ϵ_{Hf} (Moyen and Martin, 2012) that gradually evolves towards negative ϵ_{Hf} values. Archean trondhjemite, tonalite, and granodiorite (TTG) grey gneisses have ^{176}Lu/^{177}Lu ratios that remain nearly constant throughout the Archaean and are close to the *chondritic* value of 0.0336 (Bouvier et al., 2008; Moyen and Martin, 2012). These TTGs apparently developed as a result of several episodes of differentiation from magmas derived from an undepleted part of the mantle. One possible model for this involves the extraction of thick, mafic oceanic plateaus, which can be subducted and undergo further chemical differentiation after 3.0 Ga. However, our model is not necessarily bound to this assumption, especially as Guitreau et al. (2012) emphasised that ^{176}Lu/^{177}Lu ratios have only varied between 0.032 and 0.038 during the last 3800 Ma and fluctuate around the *chondritic* value of 0.0336. This suggests that the continental crust was derived from the primitive mantle as a result of several stages of differentiation, with the primitive mantle originally derived from a *chondritic* primordial Earth.

The conflict between observations and the interpretations of Boyet and Carlson (2005, 2006) and the *novel* arguments for a chondritic origin of the Earth can possibly be resolved. Graham (2002) used the low ^{129}Xe/^{130}Xe ratios of ocean island basalts (OIBs) to suggest the presence of an undegassed lower mantle reservoir. In addition, Mukhopadhyay (2012) demonstrated that the low ^{129}Xe/^{130}Xe values of OIBs cannot be explained by mixing atmospheric Xe with Xe derived from mid-oceanic ridge basalts (MORBs). He demonstrated that the He, Ne, Ar, and Xe compositions of Icelandic rocks reflect differences in the concentrations of chemical elements and ^{20}Ne/^{22}Ne ratios between MORBs and OIBs. The fact that ^{129}Xe is produced from the radioactive decay of ^{129}I, combined with ^{129}I becoming extinct at about 100 Ma after $\tau = 4567.6$ Ma, means that the MORB and OIB mantle sources must have been separated by differentiation before about 4470 Ma, with only limited subsequent mixing. This suggests that the primordial mantle *had* a chondritic Nb/U ratio, with the EDR having a superchondritic Nb/U and the hidden EER having a subchondritic Nb/U (Graham, 2010), with the Baffin Island mantle possibly representing the EDR. Uniting these ideas suggests that the sub-case IIb outlined above may be plausible. There are also different models for the generation and maintenance of the effective isolation of the deep EER and/or similar reservoirs over geologic time. For example, Lee et al. (2010) suggested that liquids are denser than the corresponding PREM densities in the magma ocean at pressures between 10 and 15 GPa, forming a layer around the present-day 410 km discontinuity (cf. Dziewonski and Anderson, 1981). At lower pressures, low-density melts could rise to the surface and form a preliminary crust, although the layer between 10 and 15 GPa would founder and would form a new layer above the CMB. This layer would be *enriched* in incompatible heat-producing elements, in noble gases (^{3}He, ^{22}Ne, and ^{40}Ar), and CO_2. The $CO_2/^3$He ratio of this layer would be approximately constant because the observed ratios in both MORBs and OIBs are not considerably different to each other (Otting and Zähringer, 1967). However, this is only one possible explanation for the existence of an apparently primordial part of the lower mantle.

C4. Heat production: what concentrations of U, Th, and K should we use in our model?

Chondritic Earth models suggest that refractory lithophile elements are present at chondritic abundances in the bulk silicate Earth (BSE) (Jagoutz et al., 1979; McCulloch and Bennett, 1994; McDonough and Sun, 1995; Palme and O'Neill, 2003). Hofmann (2003) described a typical sequence of chemical differentiation based on a chondritic mantle, where continental growth removes elements with larger ionic radii from the BSE, leaving behind a depleted region of the mantle (DM). Depending on the assumed abundances of the heat-producing elements, the DM ranges from 30% to 80% (Hofmann, 2003) or 30% to 60% of the mantle's mass (Bennett, 2003). Table 1 in the main text provides an example of the order of magnitude differences in the concentrations of elements within these geochemical reservoirs, although we used the internally consistent data presented by McCulloch and Bennett (1994) in our modelling. The 250 ppm K present in the BSE, as recorded in column McC & B of Table 1, was suggested by Jochum et al. (1983), who proposed a K/U ratio of 12,000, whereas column McC & B of Table 1 yields a nominal K/U value of 12,315.

The EDR proposed by Boyet and Carlson (2006) suggests that a K abundance of 160 ppm is possible, with no or nearly no ^{40}Ar necessary in the present-day mantle. In contrast, the 250 ppm K of the chondritic model requires about 50% of the ^{40}Ar to be stored in the deep mantle. In addition, the abundance of U and Th in the EDR would have to be only 60% of the chondritic abundance, with the bulk heat production due to the radioactive decay of U, Th and K being only 12 TW. However, measurement-based estimates of the present-day total surface heat flow range from 44 to 47 TW (Pollack et al., 1993; Davies and Davies, 2010). This poses a major problem to the EDR hypotheses of scenarios I and IIa. This suggests that primordial heat values are considerably larger than was previously thought.

However, there are a number of facts that argue against scenarios I and IIa. An ideal numerical model of the mantle's evolution should start at an initial age of $\tau = 4567.6\,\mathrm{Ma}$ (Jacobsen et al., 2008; Carlson and Boyet, 2009). Ryder (2002) presented a plot of the number of lunar craters with diameters greater than 1 km versus age in Ga, yielding an exponentially decreasing curve that can be extrapolated back to $\tau > 4100\,\mathrm{Ma}$. This in turn yields significantly higher primordial heat values for the Moon and therefore also the Earth, primarily as the intense and continuous meteoritic bombardment recorded by the Moon would also have affected the Earth. However, the Moon has a largely anorthositic crust of age $\tau = 4456\,\mathrm{Ma}$, meaning that the continuous meteoritic bombardment hypothesis is invalid. As such, we assume that a late heavy bombardment (LHB) took place between 3950 and 3870 Ma. In addition, it is likely that the Earth had a magma ocean at 4567.6 Ma, although this is inconsistent with research by Harrison et al. (2005), who obtained $^{176}\mathrm{Hf}/^{177}\mathrm{Hf}$ initial ratios for 4010 to 4370 Ma detrital zircons. Harrison et al. (2008) continued these investigations using concurrent Lu–Hf and $^{207}\mathrm{Pb}/^{206}\mathrm{Pb}$ analyses, yielding ages between 4560 and 4200 Ma for the extraction of the protoliths that formed these zircons. Little previous research suggested that the Earth's crust formed at such an early stage. Our model assumes a somewhat conservative maximum crustal age of 4490 Ma. Harrison et al. (2008) concluded that a $\mathrm{SiO_2}$-enriched crust had begun to form by 4350 Ma. Iizuka et al. (2006) also obtained zircon U–Pb ages of around 4200 Ma in a 3900 Ma granitic rock in the Acasta Gneiss Complex in Canada, and Blichert-Toft and Albarède (2008) analysed 63 zircons from the Jack Hills of Australia that crystallized 4100±100 Ma ago and were derived from a 4300 to 4360 Ma protolith. This research suggests a relatively cool early Earth *before* the LHB, thereby supporting the earlier estimates of the initial heat of the Earth.

However, if primordial heat values are considerably lower than assumed then the EDR hypothesis (variants I or II a) suggests that initial K abundances were higher than is currently the case for the EDR. This possibility is consistent with research by Marty (2012), who concluded that the H, N, Ne, and Ar isotopic compositions of the Earth can be explained by mixing chondritic and solar compositional end-members with a significant proportion of the $^{40}\mathrm{Ar}$ generated by the decay of $^{40}\mathrm{K}$ stored in the present-day silicate Earth. A recent estimate of K/U ratios is given by Arevalo et al.

(2009), who suggested K/U ratios of 19 000 ± 2600 for the DM, 11 900 ± 2200 for OIBs, 13 000 ± 3000 for the continental crust, and 13 800 ±2600 for the BSE, with the BSE also containing 280±60 ppm K. These values agree with the estimated K abundances of Jochum et al. (1983) and Table 1. Taking data from McCulloch and Bennett (1994) or from column McC & B of Table 1 yields a K/U(DM) ratio of 16 700, a K/U(continental crust) ratio of 10 100, and a K/U(BSE) ratio of 12 300, with the OIB ratio not specified. These numbers are within the limits of uncertainty of Arevalo et al. (2009). Improving on the model of Walzer and Hendel (2008), we also incorporated a number of other modifications to our mantle convection model, including the dependence of melting temperature (T_m) on water abundance. This in turn means that shear viscosity (η) is also dependent on water abundance and means that chemical differentiation started earlier. We also prescind from the initially intended revision of the assumed U, Th, and K abundances in order to obtain a better physical comprehension of our model by comparison of the solutions of our system of equations (cf. Appendix A) with the results of Walzer and Hendel (2008), especially as all of the values are within error of each other (i.e., we use the three columns McC & B in Table 1 within the main body of this paper).

Geoneutrino measurements by Gando et al. (2011) provide evidence of still higher U and Th concentrations within the Earth's mantle, suggesting that the mantle should also contain high concentrations of K. These new results argue against assuming that the K abundance of the BSE is only half as high as is needed to obtain a virtually ^{40}Ar-free present-day mantle. These data are also inconsistent with variants I and IIa of the EDR hypothesis.

However, Stracke et al. (2011) presented new Hf and Nd isotopic data for clinopyroxene from peridotites from the Gakkel and Southwest Indian ridges and the southern Atlantic. These data suggest that the average DM is considerably more depleted than is indicated by models derived from conventional MORB investigations. The residual peridotite within the DM often records a multi-stage depletion history, indicating that the DM is *not* homogeneous (as was previously suggested) but commonly contains large ultra-depleted domains that are similar to the Gakkel Ridge peridotites. In the conventional model of O'Nions et al. (1979) a mass fraction of DM of about 50% of the total mantle was estimated. This contrasts sharply with the findings of Stracke et al. (2011), who suggested that the present-day mantle contains only about 20% DM. This result also contrasts with the low-K-abundance model and the EDR proposal (variants I and IIa outlined above). Overall, for geochemical and numerical reasons we continue to use the conventional concentrations of incompatible elements according to McCulloch and Bennett (1994), primarily as these values lie within the range of compositions suggested by the most recent geochemical models. All of our physical assumptions have been updated. In addition, using conventional geochemical assumptions such as those of McCulloch and Bennett (1994) and McDonough and Sun (1995) does not cause any numerical problems in our modelling. In contrast, the use of variants I and IIa, which incorporate large assumed primordial temperature gradients at the CMB, causes numerical breakdowns.

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