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Key Points:

- We develop an array-based iterative method to measure *SmKS*-*SKKS* ($m = 3$ – 5) differential travel times
- Three-dimensional mantle structure effects must be considered in studies of *SmKS* differential travel times
- Our measurements support a low V_p at the top of outer core

Supporting Information:

- Supporting Information S1

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Array-Based Iterative Measurements of *SmKS* Travel Times and Their Constraints on Outermost Core Structure

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Abstract Vigorous convection in Earth's outer core led to the suggestion that it is chemically homogeneous. However, there is increasing seismic evidence for structural complexities close to the outer core's upper and lower boundaries. Both body waves and normal mode data have been used to estimate a P wave velocity, V_p , at the top of the outer core (the E' layer), which is lower than that in the Preliminary Reference Earth Model. However, these low V_p models do not agree on the form of this velocity anomaly. One reason for this is the difficulty in retrieving and measuring *SmKS* arrival times. To address this issue, we propose a novel approach using data from seismic arrays to iteratively measure *SmKS*-*SKKS* differential travel times. This approach extracts individual *SmKS* signal from mixed waveforms of the *SmKS* series, allowing us to reliably measure differential travel times. We successfully use this method to measure *SmKS* time delays from earthquakes in the Fiji-Tonga and Vanuatu subduction zones. *SmKS* time delays are measured by waveform cross correlation between *SmKS* and *SKKS*, and the cross-correlation coefficient allows us to access measurement quality. We also apply this iterative scheme to synthetic *SmKS* seismograms to investigate the 3-D mantle structure's effects. The mantle structure corrections are not negligible for our data, and neglecting them could bias the V_p estimation of uppermost outer core. After mantle structure corrections, we can still see substantial time delays of *S3KS*, *S4KS*, and *S5KS*, supporting a low V_p at the top of Earth's outer core.

1. Introduction

The liquid outer core in the Earth plays a critical role in the geodynamo and in thermochemical interactions between the mantle and core. Seismic studies can provide important constraints on the physical properties of the core and therefore improve our understanding of the composition and state of the core (Hirose et al., 2013). Due to vigorous convection, the bulk of the outer core is believed to be well mixed and therefore chemically homogeneous (Stevenson, 1987). However, there is increasing seismic evidence for structural complexities close to its top and bottom boundaries. A stratified layer with a lower V_p gradient than the Preliminary Reference Earth Model (PREM; Dziewonski & Anderson, 1981), labeled the F layer, has been documented using body seismic wave observations (Ohtaki & Kaneshima, 2015; Souriau & Poupinet, 1991b; Song & Helmberger, 1995; Zou et al., 2008). Another stratified layer, the E' layer, is hypothesized to exist at the top of outer core, and its properties may be constrained by geomagnetic secular variations (Buffett, 2014; Gubbins, 2007), but the seismic evidence, especially *SmKS* differential arrival times, for this layer is contradictory and controversial (e.g., Alexandrakos & Eaton, 2010; Eaton & Kendall, 2006; Helffrich & Kaneshima, 2010; Kaneshima & Helffrich, 2013).

SmKS waves ($m = 1, 2, 3, \dots$) travel as S waves in the mantle, are converted to compressional waves entering the outer core, reflected $m - 1$ times on the underside of the core-mantle boundary (CMB), and reconvert to S waves to travel through the mantle (Figure 1a). *SmKS* waves are sensitive to the structure of outer core, and their arrival times have been used to investigate V_p (compressional wave velocity) in the shallow outer core (Choy, 1977). *SKS* absolute arrival times have a large scatter, especially due to 3-D mantle structure (e.g., Garnero et al., 2016), which results in large uncertainties in their constraints on outer core structure. *SKKS* and *SKS* have similar raypaths near the source, so their differential arrival times can partially remove the source effects and constrain the V_p of shallow outer core better. Hales and Roberts (1971) compiled *SKKS*-*SKS* differential arrival times and found a low V_p in the outermost core. However, the reliability of

Figure 1. Raypaths of *SmKS* waves and one example of *SmKS* waveforms. (a) Raypaths of *SmKS*. The red star is an earthquake, and the blue triangle represents a seismic station. The green line shows the raypath of *SKKS* traveling in the outer core. The black lines are raypaths of *SmKS* ($m = 3, 4$, and 5) and sections of *SKKS* raypath traveling in the mantle and crust. (b) A band-pass-filtered (bp 0.05–0.7 Hz) seismogram of *SmKS* data from station ASSE with an epicentral distance of 150.5° from Event #110729 (Table S1 in the supporting information). Time zero is the *SKKS* arrival predicted by PREM. The predicted arrival time of *S3KS* is 39 s after *SKKS*. *S4KS* arrives at 52 s, and *S5KS* is only 5 s after the *S4KS*.

this study is reduced by the uncorrected phase shifting between *SKS* and *SKKS* (Choy & Richards, 1975; Choy, 1977).

Although the raypaths of *SKS* and *SKKS* are close to each other near the source, they diverge further in the lower mantle, where lateral heterogeneities could affect their different travel times (Garnero et al., 1988; Souriau & Poupinet, 1991a). Compared to *SKKS* and *SKS*, *SmKS* and $S(m-1)KS$ with $m > 2$, for example, *S3KS-SKKS*, have closer raypaths (Figure 1a), and therefore, their differential arrival times are less affected by 3-D mantle structures. With the high-quality seismic data accumulated in the last few decades, many more observations of *SmKS* ($m \geq 2$) waves have been reported and their differential travel times have been used to investigate the stratification of the top outer core. However, the conclusions of various studies are not consistent. For example, Alexandrakis and Eaton (2007) exploited the Empirical Transfer Function technique to precisely measure *SmKS* differential travel times and found no evidence for stratification, consistent with some other *SmKS* studies (e.g., Alexandrakis & Eaton, 2010; Souriau & Poupinet, 1991a). In contrast, other reports support a layer with lower V_p than that of PREM in the outermost core (e.g., Eaton & Kendall, 2006; Kaneshima & Helffrich, 2013; Garnero et al., 1993; Helffrich & Kaneshima, 2010; Kaneshima & Matsuzawa, 2015; Kaneshima, 2018; Kaneshima & Helffrich, 2013; Tanaka, 2004, 2007; Tang et al., 2015), although the thickness and amplitude of the V_p anomaly varies from one study to another.

There are at least two reasons for the preceding contradictory results. The first one is the difficulty in extracting each individual *SmKS* phase and precisely measuring the differential arrival times. For high orders $m \geq 3$, *SmKS* series constitute a whispering-gallery mode and consecutive *SmKS* phases have very close arrival times (e.g., *S4KS* and *S5KS* in Figure 1a), which makes separating consecutive *SmKS* waveforms difficult. Another problem is contamination from lateral heterogeneities in mantle structure. Although raypaths of *SmKS* and $S(m-1)KS$ ($m > 2$) series are closer to each other than that of *SKS* and *SKKS*, there are still differences in

the mantle, especially the heterogeneous D'' region (Garnero & Helmberger, 1995). These mantle heterogeneities could cause large uncertainty or bias in the differential arrival time measurements made using individual seismograms (Garnero et al., 1993) or small-aperture arrays (Eaton & Kendall, 2006). Stacking of data from large-scale arrays (Helffrich & Kaneshima, 2010; Kaneshima & Helffrich, 2013; Kaneshima & Matsuzawa, 2015; Kaneshima, 2018) or global networks (Alexandrakis & Eaton, 2010) tends to average out perturbations due to mantle heterogeneities and therefore mitigate the possible bias. Alternatively, the bias can be evaluated using ray theory (e.g., Helffrich & Kaneshima, 2010; Kaneshima & Helffrich, 2013; Kaneshima & Matsuzawa, 2015; Kaneshima, 2018) or sophisticated waveform modeling (Tanaka, 2004, 2007), based on either known 3-D mantle tomography model or hypothesized structure. Combining array stacking and accurate 3-D mantle corrections would be an optimal solution to suppress 3-D mantle effects, which has not been reported before.

To ameliorate these problems, we develop an iterative method to separate individual *SmKS* phases from the *SmKS* wave train in array data and use normalized cross correlation (CC) to measure the differential travel times between *SmKS* ($m = 3, 4$, and 5) and *SKKS*. We carefully select good quality data to successfully obtain each *SmKS* phase. The iterative method provides us with accurate waveform-based measurements of differential arrival times and important information to assess the measurement quality. We use two methods, ray theory and the spectral element method (SEM), to investigate the effects of lateral heterogeneities in the mantle, using the 3-D tomography model S40RTS (Ritsema et al., 2011), and also assess the effect of choosing a different mantle model (S362ANI Kustowski et al., 2008). The measured differential arrival times, after correction for 3-D mantle structure effects, are compared to the predictions of body wave-derived model KHMOC (Kaneshima & Helffrich, 2013) and normal-mode constrained model EPOC (Irving et al., 2018).

Figure 2. Map and histogram of clear *SmKS* data. (a) Map of good *SKKS* data ($\text{SNR} \geq 2.0$) from the 11 earthquakes. The blue triangles and red stars show the stations and earthquakes, respectively. The lines connecting stations and earthquakes are raypaths of *SKKS*. The green lines show the raypaths of *SKKS* traveling in the outer core from Event #110729. (b) Histogram of *SKKS* data in (a). The blue bar portions correspond to *SKKS* data with high $\text{SNR} \geq 2.0$, and the red bar portions show the ones with $\text{SNR} < 2.0$.

2. Data

We collected more than 320,000 seismograms from global stations from 500 earthquakes in the subduction zones of Fiji-Tonga, Vanuatu, New Britain, and Solomon with depths ≥ 150 km and $M_w \geq 5.5$ (Global Centroid-Moment Tensor catalog, Ekström et al., 2012) in the period 2000–2016 (supporting information Figure S1). We select events with depths ≥ 150 km to avoid contamination from depth phases *sSmKS* ($m \geq 2$). The seismograms have a distance range of 120–180°, where waveforms *SmKS* ($m \geq 2$) are readily observed.

We remove instrument responses and rotate the two horizontal components to get the radial displacement, on which *SmKS* primarily appears. Then a band-pass filter (0.05–0.7 Hz) is applied to the data with

Figure 3. *SmKS* data from stations in Europe from Event #071016. (a) An example of waveforms with poor *SKKS* with $\text{SNR} < 2.0$ (bottom panel), good *SKKS* with $\text{SNR} \geq 2.0$ only (middle panel), and high SNRs for both *SmKS* ($m = 3$ and higher) and *SKKS* (top panel). The SNR of *SKKS* is defined as the peak-to-peak amplitude ratio of *SKKS* (20 s before to 50 s after the *SKKS* arrival predicted by PREM) to that of the noise (70 to 20 s before the *SKKS* arrival). Similarly, the SNR of *SmKS* ($m \geq 3$) is obtained by measuring *SmKS* signals (0 to 50 s after *S3KS* arrival) and the associated noise (50 to 85 s after *S3KS* arrival). The time zero is the *SKKS* arrival predicted by PREM. (b) Map of stations in Europe from Event #071016. Stations with noisy *SKKS*, good *SKKS* only, and high SNRs for both *SmKS* ($m = 3$ and higher) and *SKKS* are shown as yellow, green, and blue triangles, respectively.

signal-to-noise ratio (SNR) computation. From these 500 earthquakes, we find 11 events with a large number of good observations of *SKKS* (Figure 2a). Here, good observation means SNR larger than 2, a large number means 100 or more seismograms, and we carefully inspect the data to rule out any possible contamination from small local earthquakes. The SNR is defined as the peak-to-peak amplitude ratio of *SKKS* to noise. We measure *SKKS* amplitude in a time window between 20 s before and 50 s after the *SKKS* arrival time predicted by PREM (Figure 3a). The time window of noise is taken between 70 and 20 s before the *SKKS* arrival. There are a total of 3,741 radial components from these 11 events, and 2,535 of them have $\text{SNR}_{\text{SKKS}} > 2.0$ (Figure 2b). Limited by the geographic distribution of seismic stations, most of these clear *SKKS* data are from stations in Europe with a distance range of 140–160° and their raypaths sample the northeastern Pacific, Asia, and Europe.

Following previous studies (e.g., Alexandrakakis & Eaton, 2010; Eaton & Kendall, 2006; Helffrich & Kaneshima, 2010; Kaneshima, 2018; Tanaka, 2004), we use *SKKS* as a reference phase to investigate the arrivals of *SmKS* ($m > 2$), so clear *SmKS* ($m > 2$) signals are also important for high-quality measurements. We compute SNR of *SmKS* ($m > 2$), and only use the data with clear *SmKS* ($\text{SNR}_{\text{SmKS}} \geq 2.0$; see Figure 3). In contrast to the SNR_{SKKS} computation, we take the noise window starting after the predicted *S2KS* arrival time for SNR_{SmKS} (by 100 s) and some *SmKS* coda waves are included in this time window. Thus, the data with strong *SmKS* coda due to significant unwanted source and wave propagation complexities would have low SNR_{SmKS} and therefore be discarded. Then, we use the method described in section 3 to measure these data with clear *SmKS* ($m \geq 2$). Most of our clear data are from Europe, and our array-based method needs a number of records to form an array, so here we focus on stations in Europe and North Africa to investigate the *SmKS* arrivals.

3. Array-Based Iterative Method to Measure *SmKS*-*SKKS* Differential Arrival Times

3.1. Workflow of the Array-Based Iterative Method

SmKS ($m \geq 2$) series travel in the mantle and upper outer core, so their arrivals are sensitive to the V_s in the mantle and V_p in the outer core. The raypaths of *SKKS* and *SmKS* ($m > 2$) are close to each other in the mantle and further apart in the outer core (Figure 1a), so taking arrival time differences between *SKKS* and *SmKS* ($m > 2$), $t_{\text{SmKS}} - t_{\text{SKKS}}$, instead of absolute travel time, can significantly reduce the effects

Figure 4. Workflow of the array-based iterative method.

ence phase *SKKS* is assumed to be perfectly coherent with each *SmKS* ($m > 2$) waveform after the phase shift is applied and only two unknown parameters, *SmKS* arrival time anomalies and *SmKS*/*SKKS* amplitude ratios, are measured in each iteration. We note that another alternative measuring strategy would be attempting to measure *SmKS*- $S(m-1)$ KS (i.e., *S3KS*-*SKKS*, *S4KS*-*S3KS*, and *S5KS*-*S4KS*), which have even closer raypaths than those of *SmKS*-*SKKS* ($m = 3, 4$, and 5). However, this strategy suffers from the problem of weak and noisy reference phases *S3KS* and *S4KS*, which would affect the performance of our method. Thus, we choose the clearer *SKKS* waveforms as the reference phase.

This workflow of our iterative method is composed of data preparation and then iterative measuring (Figure 4). As described in subsection 2, we set an SNR threshold of 2 for both *SKKS* and *SmKS* ($m > 2$) to obtain good quality data. Following Helffrich and Kaneshima (2010) and Kaneshima and Helffrich (2013), we divide the clear *SmKS* data from the same event into several bins and stack traces in each bin to further improve the SNR (an example of one bin is shown in Figure 5). Before stacking the traces, two steps of CC are carried out on *SKKS* waveforms to align the data. In the first step of CC, we choose one typical trace (i.e., station with the median distance of the bin) as a template (e.g., black line in Figure 5b) and compute CC of *SKKS* between this template and other traces in this bin with shifting times. Then these traces are aligned on the time with the maximum CC values. In the next step, we stack the aligned *SKKS* with normalized amplitudes to form a new template (e.g., red line in Figure 5b) and then repeat the CC processing to align the *SmKS* data again (Figure 5c). The time window of *SKKS* used in CC is 5 s before and 30 s after the arrival time of *SKKS* and the maximum allowed time shift is 5 s. Data with maximum CC coefficients lower than 0.8 are not used in the following iterative measuring, because their low waveform similarities, due to complex site structure or/and instrumental issues, could decrease the quality of stacking and affect the measurements. In these two steps of alignment, the shifted times are primarily due to 3-D structures near the stations, source mislocation, and/or clock time errors and these factors are shared by *SKKS* and *SmKS* ($m > 2$). Thus, shifting the traces are not expected to significantly affect the measurements of differential arrival times.

Next, we use these aligned *SmKS* data to iteratively retrieve individual *SmKS* phases, measure differential travel time anomalies, and assess quality of each measurement. In the first iteration (“iteration1” in Figure 4), we stack the data in a bin and use three CC processes to measure *S3KS*, *S4KS*, and *S5KS* one

of 3-D Vs structure in the mantle and improve the constrains on the *Vp* in outer core. On the other hand, these spatially close raypaths result in small time separations between consecutive *SmKS* signals, which can make identifying individual *SmKS* phase and measuring its arrival time difficult. For example, the arrival time difference between *S3KS* and *S4KS* at station ASSE from Event #110729 is only 13 s (Figure 1a). The difference between *S4KS* and *S5KS* is even smaller and their waveforms are mixed with each other. Many previous efforts have been made to retrieve individual *SmKS* phase and accurately measure their arrival times (e.g., Eaton & Kendall, 2006; Helffrich & Kaneshima, 2010; Kaneshima & Helffrich, 2013). In particular, array stacking techniques have been used to analyze slownesses and arrival times of *SmKS* signals (e.g., Eaton & Kendall, 2006; Helffrich & Kaneshima, 2010; Kaneshima & Helffrich, 2013). Here, we take the advantage of the large number of stations with good data to form one or more arrays or bins and develop an iterative method to retrieve individual *SmKS* and measure their arrival times. This iterative strategy has been used to extract direct *S* waves and CMB reflected *ScS* waves (Yu et al., 2012).

Arrivals in the *SmKS* series share many factors, such as source time function, 3-D wave propagation effects, and site responses, due to their similar raypaths in the crust and mantle. Although their raypaths diverge further in the outer core, the outer core is believed to be highly laterally homogeneous. Thus, *SKKS* and *SmKS* ($m > 2$) usually have very coherent waveforms (after a $\pi(m-2)/2$ phase shift is applied to *SmKS* with $m = 3, 4$, and 5). This property helps us significantly simplify the problem and separate individual *SmKS* waveform. In our iterative method, the refer-

Figure 5. An example of aligning *SKKS* by two steps of cross correlation. (a) A map of a bin of stations with clear *SmKS* from Event #071016. The other stations with clear *SmKS* are shown in Figure 2b. (b) The *SKKS* waveforms from the reference station GRA2 (upper trace, epicentral distance of 154.04°) and stacked *SKKS* after alignment by CC with GRA2 (lower panel). (c) Distance profile of *SmKS* data (0.05–0.7 Hz) aligned on *SKKS* by two steps of CC. The corresponding stations are shown in (a). The time zero is the *SKKS* arrival. The other red dashed lines are the *SmKS* ($m = 3$ –5) arrivals predicted by PREM.

by one. For *S3KS* measurement, we stack *S3KS* using $t_{S3KS} - t_{SKKS}$ predicted by PREM (e.g., see the second red dashed line Figure 5c), apply the Hilbert transform on them to correct the 90° phase shift, and then compute CC between stacked *SKKS* and *S3KS* to get *S3KS*/*SKKS* amplitude ratio and time delay of *S3KS*. Then we cut out the *SKKS* waveform at each station, scale them using the previously measured *S3KS*/*SKKS* amplitude ratio, and apply the phase shift to get *S3KS* waveform estimation. This estimated *S3KS* is subtracted from the data to retrieve a “clean” *S4KS*, and then a similar stack-CC processing is applied on the retrieved *S4KS* for measurement. Once *S3KS* and *S4KS* have been measured, we can estimate both *S3KS* and *S4KS*, remove them in the data, and then measure *S5KS*. After *iteration1*, we obtain initial estimations of *SmKS*/*S2KS* ($m = 3, 4$, and 5) amplitude ratios and their time delays. In the next iteration, these information are used to retrieve the target *SmKS* and more accurately measure them. This iteration is repeated until the measurements are convergent.

This array-based iterative method uses good quality data and has the advantages of enhancing SNR by stacking and retrieving target *SmKS* signals well by removing other *SmKS* interfering signals. Note that we use theoretical slowness derived from PREM to stack array data, because V_p anomaly in the uppermost outer core only causes small slowness deviation and slowness measurements could have large uncertainties. A large slowness anomaly would result in less coherent stacking, which would be reflected in the CC coefficient. In the first step of data preparation, we set strict criteria to rule out the data with potential issues that might affect the validity of our method. For example, the requirement of $SNR_{SKKS} \geq 2.0$ allows us discard the data with high noise before *SKKS*. In addition to that, the other two thresholds of $SNR_{SmKS} \geq 2.0$ and $CC \geq 0.8$ rule out more bad quality data (e.g., complex *SmKS* waveforms and/or strong *SmKS* coda waves due to 3-D heterogeneity or source or station structures). Stacking the data with high CC value further increases SNR and extracting individual *SmKS* phase from mixed signals allows us reduce uncertainties in

measurements. More importantly, this method provides us two critical parameters to assess qualities of measurements. The most important parameter is the CC values between $S2KS$ and target $SmKS$ ($S3KS$, $S4KS$, and $S5KS$). A low CC value means a bad quality measurement, and we should either discard it, or be careful when using it. Low CC values could be due to a failure of the assumptions we made, weak target signals (e.g., near the nodal plane of radiation pattern of earthquake), insufficient number of traces in a bin, and so on. In addition to CC values, the amplitude information is also helpful to assess measurement quality. More details are discussed in section 5.

Uncertainty of differential arrival time for each bin is estimated by bootstrapping (Efron & Tibshirani, 1991), which reflects the variance in the bin. For each bin, we randomly select N seismograms, with replacement, from the original N seismograms and measure the differential arrival times. This process is repeated 300 times, and we compute the standard deviation of these 300 measurements as an estimation of variance in that bin.

In next section, we demonstrate the validation of our method by testing synthetic seismograms and then apply it to data.

3.2. Synthetic Tests

In this subsection, we cut real $SKKS$ waveforms from data, use them to make $SmKS$ ($m = 2, 3, 4$, and 5) synthetics, and then validate our iterative method. Figure 5c shows $SmKS$ data of a bin from Event #071016. We cut and taper the $SKKS$ waveforms from 0 to 40 s as input to generate $S3KS$, $S4KS$, and $S5KS$ (Figure 6). $S3KS$ is formed by scaling the input signals with a prescribed $S3KS/SKKS$ amplitude ratio of 0.42, applying a 90° phase shift and a prescribed time shift, which is 1.13 s greater than to the PREM $S3KS$ - $SKKS$ differential arrival time. Similarly, $S4KS$ and $S5KS$ are made with different amplitude ratios and time delays. Then, complete $SmKS$ synthetic seismograms are generated by summing $SKKS$, $S3KS$, $S4KS$, and $S5KS$.

Then we apply our iterative method to these synthetic seismograms and check its validity. In the step of searching maximum CC values, we take a time window of 0–30 s after the target $SmKS$ arrival time and the maximum allowed time shift is 5 s. In previous studies, the time delays of $SmKS$ ($m = 3, 4$, or 5) are less than 5 s and most of them are less than 3 s (e.g., Eaton & Kendall, 2006; Helffrich & Kaneshima, 2010; Kaneshima & Helffrich, 2013).

Figure 7 shows the measurements from the first five iterations. We can see that both amplitude ratios and time delays are successfully retrieved and the CC values for $S3KS$, $S4KS$, and $S5KS$ are higher than 0.95 after the second iteration. In the first iteration, there are some differences between the measured results and true values. For example, the measured time delay of $S4KS$ is ~ 2.15 s, which is ~ 0.1 s smaller than the input 2.25 s. The CC value for $S3KS$ measurement, $CC_{3,2}$, is 0.88, lower than $CC_{4,2} = 0.92$ for $S4KS$ and $CC_{4,2} = 0.92$ for $S5KS$, because the $S3KS$ measurement is affected by the presence of $S4KS$ and $S5KS$ signals. In the second iteration, the CC values are significantly increased and the measurements are close to the true values. The measurements become almost constant in the next three iterations, showing they reach convergence. After the first two iterations, waveforms are successfully retrieved and the time delays are accurately measured (e.g., see the waveform CCs between $SKKS$ and $S5KS$ in Figure S2). These results demonstrate the validation of our method. Of course, real data may be more complex than the synthetic $SmKS$ here, for example, different noise signals may be present in data, and therefore, measurement quality might be not as good as in these synthetic tests. However, CC values indicate this complexity, demonstrating their importance.

3.3. Correcting 3-D Mantle Structure Effects

Because the raypaths of $SmKS$ ($m = 2$ – 5) are close to each other in the mantle, many previous studies assume that the effects of 3-D mantle structures are the same for $SKKS$ and $SmKS$ ($m > 2$). Thus, the measured time delays of $SmKS$ ($m > 2$) are only due to the V_p anomalies in the top outer core. However, we know that the raypaths between $SKKS$ and $SmKS$ ($m > 2$) are not exactly the same and the 3-D mantle structures must affect the arrival time difference between $SKKS$ and $SmKS$ ($m > 2$). Kaneshima and Matsuzawa (2015) used ray theory to investigate these mantle effects at receiver side and source side. At the receiver side, they found that the mantle effects on $dt_{3,2}$ are much less than 0.4 s. However, the presence of a Large Low Shear Velocity Province beneath the Pacific could cause some time delays of $SmKS$ ($m \geq 2$) and affect the measurements.

Figure 6. Making *SmKS* ($m = 2-5$) synthetics using the *SKKS* data from Event #071016. The upper left figure shows the cutout and tapered *SKKS* waveforms (complete data shown in Figure 5c). Each subfigure in the lower panel corresponds to the synthetics of individual *SmKS* phase. We take the tapered *SKKS* data (upper left figure), scale them using given amplitude ratios, and apply the corresponding phase shift and time shift to form each *SmKS* phase. The *S3KS*/*SKKS* amplitude ratio is given as 0.42, and its time delay is 1.13 s. For *S4KS*, the amplitude ratio is 0.31 and the time delay is 2.25 s. For *S5KS*, the amplitude ratio is 0.14, and the time delay is 2.39 s. These *SmKS* phases are added together to form the complete synthetics of *SmKS* series (upper right figure). The dashed red line at the time zero in each figure is the *SKKS* arrival, the same as Figure 5c. The other red dashed lines are the *SmKS* ($m = 3-5$) arrivals predicted by PREM.

To investigate 3-D mantle effects, we use two different methods, ray theory and SEM, to compute the travel time delays of *SmKS* and compare their differences. We use SPEC-FEM3D_globe to compute synthetic seismograms and evaluate the 3-D mantle effects present in the tomography models S40RTS (Ritsema et al., 2011) and S362ANI (Kustowski et al., 2008). As a SEM, the SPEC-FEM3D_globe package solves the weak form of the seismic wave propagation equation and has the advantages of high accuracy, fast computation speed, handling discontinuity topography, and so on (Komatitsch & Tromp, 1999, 2002; Tromp et al., 2008).

Figure 7. A synthetic test to validate the array-based iterative method. The colored circles indicate the measured time delay in each iteration. The colored squares are the measured amplitude ratios. The upper black dashed line in each figure is the prescribed time delay and the lower black dashed line corresponds to the given amplitude ratio. Note that the time delays are relative to *SmKS-SKKS* differential arrival times predicted by PREM. The color represents the CC values between the single *SmKS* phase and transformed *SKKS* (e.g., results of *S5KS* from the first four iterations shown in Figure S2). Both time delays and amplitude ratios of *S3KS*, *S4KS*, and *S5KS* converge to the input values after five iterations and this is also reflected in the high CC coefficients.

The adjoint source technique is part of *SPECFEM3D_globe*, allowing the efficient computation of global scale sensitivity kernels of seismic signals in a given time window and frequency band (Luo et al., 2013; Tromp et al., 2008). We set the mesh parameters *NEX_XI* and *NEX_ETA* to 896, and the minimum resolved period is about 4.9 s. We use source parameters from GCMT (Global Centroid Moment Tensor, Ekström et al., 2012) and *SPECFEM3D_globe* to compute the synthetic seismogram and the *SmKS* travel time sensitivity kernels. We note that GCMT solutions do not contain detailed inversions for source duration, so we reestimate the source durations using teleseismic *P* waves from global stations. For comparison, we also compute the 3-D mantle structure corrections based on ray theory using the *S40RTS* model. To simplify the problem, we use PREM to get the raypath and compute the arrival time perturbations along that raypath. In other words, we assume that the raypath is not dramatically distorted by 3-D structures.

Figure 8 shows a depth cross section of fractional velocity anomaly, dV/V_s , from the 3-D model *S40RTS* (Ritsema et al., 2011) along the great circle connecting station GRA1 and Event #110729. The raypaths of *SmKS* ($m = 2, 3$, and 4) sample the Large Low Shear Velocity Province at the source side, where dV_s is lower than -1% . Using this 3-D V_s mantle model, we can use ray theory to compute the arrival time anomalies of *SmKS* ($m = 2, 3, 4$, and 5) along their raypaths. However, ray theory only works at infinite frequency. Indeed, seismic waves at a finite frequency are sensitive to a Fresnel zone, a region centered at its raypath. To demonstrate the Fresnel zones of *SmKS*, we use the *SPECFEM3D_globe* package (Komatitsch & Tromp, 1999) to compute sensitivity kernels of *SmKS* in the mantle and outer core.

Figure 8a shows the *SPECFEM3D_globe* synthetic seismogram at GRA1 from Event #110729 and three time windows used to compute the sensitivity kernels of *SKKS*, *S3KS*, and *S4KS*. We use the GCMT solution (Ekström et al., 2012) as the input of source parameters but reestimate its source duration (Figure S4). The *S40RTS* model is used to describe mantle heterogeneity and attenuation simulation is disabled to speed up the computation. We use 1536 CPU cores to run the SEM simulation, taking about 14 hr for forward modeling and 27 hr for each adjoint simulation. At frequency 0.05–0.2 Hz, the first Fresnel zone of *SKKS* (the green band centered at *SKKS* raypath) has a width of $\sim 18^\circ$ (~ 900 km) on the CMB and its upper boundary approaches the raypaths of *S3KS* and *S4KS* (Figure 8). The sensitivity kernels of *S3KS* and *S4KS* have similar dimensions (i.e., the width of the first Fresnel zone), but more complex patterns than *SKKS*. Compared to *SKKS*, *S3KS* and *S4KS* are more sensitive to the shallower outer core, reflected in the distribution of sensitivity kernels. The wide dimensions and complex patterns of *SmKS* sensitivity kernels in Figure 8 indicate that the 3-D mantle structure corrections based on ray theory may cause systematic biases and uncertainties. We will discuss the detailed 3-D mantle structure correction of each bin and the comparison of ray theory and SEM results in section 4.2.

Figure 8. Map and travel time sensitivity kernels of $SmKS$ ($m = 2-4$) at station GRA1 from Event #110729. (a) SEM synthetic seismogram (upper panel) and map (lower panel). The red line in the upper figure is the radial component synthetic seismogram of station GRA1 at 0.05–0.2 Hz. The triangle in the map shows the location of station GRA1 and the star is the centroid location of Event #110729. The black line in the map shows the great circle path of $SmKS$. The arrival times predicted by PREM are 1,717.0 s for $SKKS$, 1,758.5 s for $S3KS$, and 1,772.1 s for $S4KS$. The centroid time is 2.5 s, half of our reestimated duration (Table S1), after the origin time for this event. (b) Travel time sensitivity kernels of $SKKS$. Sensitivity to V_s is shown in the mantle and to V_p in the core. The red-blue colors illustrate the depth cross section of dV_s/V_s (V_s perturbation) of the 3-D model S40RTS. The green-yellow colors show the travel time sensitivity kernels of $SKKS$, and its raypath is plotted with the black line. The dashed black lines are the raypaths of $S3KS$ and $S4KS$. (c) Travel time sensitivity kernels of $S3KS$. (d) Travel time sensitivity kernels of $S4KS$.

4. Results

4.1. Measuring $SmKS$ - $S2KS$ Differential Arrival Times

We apply the iterative method to data at three frequency bands (0.05–0.2, 0.05–0.7, and 0.1–0.7 Hz) and investigate the time delays of $S3KS$, $S4KS$, and $S5KS$. For each frequency band, we compute the SNRs (SNR_{SKKS} and SNR_{SmKS}), take clear $SmKS$ data of each event to form bins (one example of Event #141101 shown in Figure S3), and apply the iterative method to each bin.

We only use data at epicentral distances greater than 140° . At shorter distances, *S3KS* arrival times are close to *SKKS* (i.e., arrival time difference smaller than 27 s) and therefore might affect quality of cut *SKKS* waveforms. Based on the number of clear *SmKS* traces and the station distribution, we divide the data from each event into several geographical bins. For example, Event #141101 provides more than 100 clear *SmKS* traces (0.05–0.2 Hz), and we divide them into five bins (see Table S2 and Figure S3). For some bins (e.g., Bin 2 from Event #010526 in Table S2), the number of clear *SmKS* traces is too few (i.e., <10) to provide reliable measurements, so we do not use the results of these bins.

At frequencies 0.05–0.2 Hz, we eventually have 24 effective bins from the 11 events (Table S2). We use the same parameters (i.e., a time window of 40 s to cut *SKKS* and 35 s for CC computation) as in synthetic testing and apply the iterative method to each bin. In the synthetic testing, the measured results are almost constant after the second iteration. Thus, here we conduct six iterations and take the results from the fifth iteration (detailed measurements listed in Table S2). For each bin, we check the results and make sure that there is no substantial difference between the fourth and fifth and six iterations. In the 24 bins, the measured *S3KS* time delay, $dt_{3,2}$, ranges from -0.03 to 2.83 s and the *S3KS/SKKS* amplitude ratios, $A_{3,2}$, are between 0.35 and 0.71. Nineteen bins have $CC_{3,2} \geq 0.90$, and most of the time delays are positive values, except Bin 1 from Event #010428. *S4KS* and *S5KS* are more difficult to retrieve and measure. This is reflected in the generally lower CC values and larger measurement scatter than *S3KS*. Figure 9 shows an example of Bin 4 from Event #141101. All the three CC values are higher than 0.94, indicating good quality measurements. For this bin, the measured time delays are 1.30 s for *S3KS*, 2.48 s for *S4KS*, and 2.59 s for *S5KS*. The median epicentral distance of this bin is 145.29° , and those time delays would indicate a slower V_p than in PREM in the topmost outer core, consistent with previous studies (e.g., Eaton & Kendall, 2006; Helffrich & Kaneshima, 2010; Kaneshima & Helffrich, 2013).

The measurement qualities are primarily indicated by their CC coefficients. In addition to CC coefficients, amplitude information is also useful to assess the measurement quality. If other factors, such as source radiation pattern, are the same, the amplitude of the *SmKS* phase decreases with its order m , due to the energy loss at each reflection on the underside of the CMB. All the measurements with good quality at 0.05–0.2 Hz follow this trend of $A_{3,2} > A_{4,2} > A_{5,2}$ (amplitude information in Table S2 and the good quality measurements are listed in Tables S3–S5).

4.2. Three-Dimensional Mantle Structure Corrections

We run SPECSEM3D_globe to obtain the synthetic seismograms corresponding to the data with good quality measurements. Here, good quality means that more than ten traces are used in a bin and $CC_{3,2} \geq 0.90$ (Table S2). Most source parameters used in the SEM simulations are from GCMT, but the source durations are replaced with our estimated values. Then we apply our iterative method to these synthetic seismograms to obtain the time delays, amplitude ratios, and corresponding CC values. For most bins, we successfully retrieve signals of *S3KS*, *S4KS*, and *S5KS* and get high CC coefficients (Table S2). For example, Figure S5 shows the measurements using synthetic seismograms corresponding to the Bin 4 from Event #141101. The CC coefficients are 0.95 for *S3KS*, 0.96 for *S4KS*, and 0.94 for *S5KS*, indicating good measurement quality. The *S3KS* time delay, $3dM_{3,2}^{SEM}$, is as large as 0.60 s and the *S4KS* time delay, $dt_{4,2}^{SEM}$, is even larger, 1.13 s. The time delays measured on the data are 1.31 s for *S3KS* and 2.48 s for *S4KS* (Table S2). Thus, 3-D mantle structure corrections are large, up to nearly half the size of the observations, and cannot be ignored for this bin. The *S3KS* measurements on synthetic seismograms of other bins are listed in Table S3 and almost all the bins have $CC_{3,2}^{SEM}$ higher than 0.95, except the Bin 1 from #010428 and Bin 1 from #140721. The *S3KS/SKKS* amplitude ratios range from 0.30 to 0.51 and the corrections to *S3KS* time delays are between -0.95 and 0.04 s. Most of the corrections have negative values, indicating that *S3KS* are delayed more than *SKKS* by the 3-D mantle structure. The results for *S4KS* and *S5KS* are listed in Tables S4 and S5.

We also use ray theory to compute 3-D mantle structure corrections for data at individual stations in each bin and take the average value to represent the correction for that bin. These corrections are close to that measured on SEM synthetic seismograms (Tables S3–S5 and Figure S6). However, large discrepancies are present for some bins. For example, the correction to *S3KS* time delay based on ray theory is 0.27 s for Bin 5 from #141101, but it is -0.22 s using SEM synthetic seismograms.

Figure 10 shows the *SmKS* ($m = 3, 4$, and 5) time delays measured on the data with high CC coefficients and the results after 3-D mantle structure corrections. Here, we require $CC_{3,2} \geq 0.90$ for a good quality of *S3KS* measurement. For *S4KS*, we only take the bins with $CC_{4,2} \geq 0.85$ and $CC_{3,2} \geq 0.90$, because a good quality

Figure 9. Time delays of *S3KS*, *S4KS*, and *S5KS* measured on Bin 4 data after five iterations from Event #141101 (0.05–0.2 Hz). (a) CC between *SKKS* and *S3KS* (after Hilbert transform and polarity inverted). The black line is the stacked *SKKS*, and the red line represents the stacked *S3KS*. The green line shows the shifted *S3KS* with the maximum CC value. The time shift between the red line and green line is 1.30 s, and the corresponding CC value is 0.94. Note that the time delay is relative to *S3KS-SKKS* differential arrival time predicted by PREM. (b) CC between *SKKS* and *S4KS* (polarity inverted). (c) CC between *SKKS* and *S5KS* (after Hilbert transform).

of *S4KS* measurement relies on a well-retrieved *S3KS*. Similarly, we require $CC_{3,2} \geq 0.90$, $CC_{4,2} \geq 0.85$, and $CC_{5,2} \geq 0.80$ for good quality of *S5KS* measurements.

Most of the bins with good qualities of $dt_{3,2}$ measurements have uncertainties smaller than 0.4 s (Table S3). It is not surprising that $dt_{4,2}$ and $dt_{5,2}$ generally show larger uncertainties than $dt_{3,2}$, due to their smaller SNR and/or incomplete separation of *SmKS* ($m = 2, 3, 4$, and 5) waveforms of our method. In spite of this, the uncertainties are still much smaller than the anomalies (Tables S4 and S5), because the bins with large errors, resulted from poor phase stripping and/or low SNRs, are discarded by the CC requirements. We note

Figure 10. *SmKS* time delays measured at 0.05–0.2 Hz. The empty squares represent the *SmKS* ($m = 3$ in a, 4 in b, and 5 in c) time delays measured on the data. Note that error bars are symmetric and in a few cases extend beyond the limits of the figure. The solid circles are *SmKS* time delays after the 3-D mantle structure corrections based on ray theory and using S40RTS model. The time delays are relative to *SmKS*-SKKS differential arrival times predicted by PREM. The solid diamonds are *SmKS* time delays after the corrections measured on the SEM synthetic seismograms made using S40RTS. The color shows the corresponding CC values measured on the data. More detailed information is displayed in Tables S3–S5. The black dashed line in each figure is the corresponding *SmKS* time delay predicted by KHOMC (Kaneshima & Helffrich, 2013). The black dotted lines show the EPOC predictions. The source depth used in the KHOMC and EPOC predictions is 150 km. (a) S3KS time delays. (b) S4KS time delays. (c) S5KS time delays.

that bootstrapping results only help us infer variance in the data set but not able to estimate systematic bias. The systematic bias could be due to strong mantle heterogeneities and source complexities and so on, which can be assessed by investigating global data from earthquakes at various places.

4.3. Comparison Between Observations and Predictions of Two 1-D Models, EPOC and KHOMC

From Figure 10, we can see that S3KS, S4KS, and S5KS time delays predicted by EPOC (Irving et al., 2018) and KHOMC (Kaneshima & Helffrich, 2013) are close to each other at distance 140–155°, where most of our data are located. The measured S3KS time delays are generally positive and consistent with the KHOMC and EPOC predictions supporting a slower V_p in the top outer core. The 3-D mantle structure corrections, using either ray theory or SEM synthetic seismograms, are primarily negative and therefore reduce the measured S3KS time delays. After the corrections, travel time anomalies are less than the EPOC predictions and they seem to fit the KHOMC predictions better than that of EPOC. The difference between these two models can be better resolved using data at distances $> 160^\circ$, where their difference is larger than 0.5 s. Unfortunately, we have only one such datum, at a distance of 167.82°, so we can not clearly distinguish between EPOC and KHOMC. For S4KS and S5KS, the 3-D mantle structure corrections are also primarily negative and they make measurements closer to the KHOMC and EPOC predictions. However, there are a few measurements dramatically departing from the EPOC and KHOMC predictions. For example, $dt_{3,2}^{SEM}$ of bin 1 from #140721 is -0.22 s while the EPOC prediction is 1.22 s. For this bin, the two types of 3-D mantle structure corrections have a large difference, 0.12 s from ray theory computation and -0.95 s from SEM synthetic seismograms. This large difference could be due to the limitation of ray theory, uncertainty in the S40RTS model, or poor performance of our method on the synthetic seismograms of this bin. The $CC_{3,2}^{SEM}$ is only 0.91, much lower than that of other bins, which indicates a poor measurement quality. However, $CC_{3,2}$ from data is a high value of 0.96 and its $dt_{3,2}^{ray}$ is close to the EPOC and KHOMC predictions. This big difference is most likely due to a large uncertainty in the 3-D mantle corrections using SEM synthetic seismograms. Some other bins, including S3KS time delays of Bin 1 from #010428 and S5KS time delays of Bin 1 from #010516, have similar issues. Note that the S5KS time delay of Bin 1 from #010516 is beyond the y axis range and not plotted in Figure 10c.

We also apply two other filters, 0.05–0.7 and 0.1–0.7 Hz, to the data and repeat the measurements. Because running SPEC3D_globe to resolve a frequency of 0.7 Hz is very computationally expensive, we only compute the 3-D mantle structure corrections using ray theory. Similar to the results at 0.05–0.2 Hz, the S3KS, S4KS, and S5KS measurements at 0.05–0.7 Hz are close to the EPOC and KHOMC predictions after the 3-D mantle structure corrections (see Figure S7).

Note that the bins shown in Figure S7 are not the same as 0.05–0.7 Hz, because the SNRs of data may change with frequency band and the measurement qualities could also be different. Comparing to 0.05–0.2 and 0.05–0.7 Hz, the number of bins with good measurement qualities is lower at 0.1–0.7 Hz, indicating lower SNRs of data and/or reduced performance of our iterative method for this high-frequency band for the data used here. Relatively long period *SmKS* waves have been stacked to investigate outermost core structure (e.g., 0.02–0.1 Hz in Tanaka, 2007). Shorter period waves have the potential to resolve finer seismic structure. However, source rupture processes and propagation effects due to lateral heterogeneities could give rise to more waveform complexities at shorter period waves reducing the waveform coherencies of *SmKS* phases and therefore affecting measurement qualities. This might explain the lower number of good quality measurements (Figures S7d–S7f) at 0.1–0.7 Hz than that at 0.05–0.2 Hz (Figure 10) and 0.05–0.7 Hz (Figures S7a–S7c).

5. Discussion

SmKS differential arrival times are sensitive to the outer core structure, but accurate measurements of differential arrival times are hampered by their mixed waveforms as a whispering-gallery mode. To extract each individual *SmKS* phase, Eaton and Kendall (2006) use *SKKS* as reference waveform and apply deconvolution to *SmKS* series to convert their waveforms into simple pulses. However, the deconvolution method either requires very high SNR and/or has reduced resolution. Here, we develop an iterative method to isolate individual *SmKS* waveforms with a high resolution. Our method keeps waveform features of each *SmKS* and therefore allows us to measure *SmKS* time delays by CC.

We use two different methods, ray theory and SEM synthetic seismograms, to compute effects of mantle heterogeneities and make these corrections to the measurements. The corrections are between –0.5 and 0.5 s for most bins, but some bins have 3-D mantle perturbations even greater than 1.0 s. Furthermore, we see big differences between the two types of corrections for some data (e.g., >1 s for S3KS-SKKS from Bin 1 from Event #140721), although they generally have positive correlation (Figure S6). We also use another 3-D mantle model S362ANI (Kustowski et al., 2008) to compute the SEM synthetic seismograms and measure the 3-D mantle structure corrections. Compared to our results using S40RTS, the corrections to S3KS-SKKS, S4KS-SKKS, and S5KS-SKKS differential arrival times using S362ANI are generally stronger. Consequently, the corrected *SmKS*-SKKS ($m = 3, 4, \text{ and } 5$) time delays become even smaller (Figures S8 and S9). Helffrich and Kaneshima (2013) used earthquakes in Fiji and Argentina to investigate *SmKS*-SKKS time delays. Their measured S3KS-SKKS time delays from Fiji are generally larger than that from Argentina. The earthquakes in our study are geographically close to Fiji and the 3-D mantle corrections to S3KS-SKKS time delays tend to reduce the S3KS-SKKS time delays (Figure 10a). Thus, the higher S3KS-SKKS time delays from events in Fiji by Helffrich & Kaneshima (2013) can be largely explained with the 3-D mantle structure. Three-dimensional mantle structure corrections should be routinely considered to reduce bias in the V_p estimation of uppermost outer core.

After correcting for 3-D mantle structure, there are still significant *SmKS*-SKKS time delays at all of the three frequency bands (Figures 10 and S7), indicating a lower V_p than PREM model in the shallow outer core. Strong locally concentrated heterogeneities, such as the previously detected Ultra Low Velocity Zones (ULVZs) at the source side of our study region (see, e.g., the compilations by Yu & Garnero, 2018), are not accurately represented in the smooth global tomography model of S40RTS and could affect the measurements. However, these ULVZ effects have been investigated by Tanaka (2007) and they are expected to be smaller than our measured time delays. In addition, such strong heterogeneities would decrease the coherencies between SKKS and *SmKS* ($m = 3, 4, \text{ and } 5$) and only the results with high CC values are selected in our method. Further, quantitative investigations will rely on better constraints on the properties and geographical distributions of ULVZ and more detailed numerical waveform modeling. Thus, we do not believe that the *SmKS*-SKKS travel time delays are solely due to ULVZs but do indeed indicate a seismically slow uppermost outer core.

Although scatter and uncertainty are present, our measurements are generally consistent with the predictions by the KHOMC and EPOC models. Assuming the outer core is homogeneous, Irving et al. (2018) use a physically consistent equation-of-state to parameterize the elastic properties of outer and carry out inversions for seismic normal mode data. This normal mode derived EPOC model shows lower V_p and higher

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density than PREM at the top of outer core. Although EPOC does not use body wave data, its fit to *SmKS* data is better than PREM (see Figure 3 in Irving et al., 2018, and Figures 10 and S7 in this study). KHOMC is derived from *SmKS* body wave travel time anomalies and has higher depth resolution than EPOC. We note that both EPOC and KHOMC models have a low V_p at the top of outer core, but they have different depth gradients of V_p . KHOMC seems to fit our results better than EPOC. For example, EPOC overpredicts most *S3KS-SKKS* time delays after 3-D mantle corrections. However, given the scatter present in our measurements, either EPOC or KHOMC fits the data well. The contrast between stratified or homogeneous structure has important implications for understanding the thermochemical status of core and the associated geodynamo. A stratified outer core would change the flow in the outer core and therefore affect the secular variation of geomagnetic field (e.g., Braginsky, 1993; Buffett, 2014; Buffett et al., 2016). However, the detailed effects of such stratification on the geodynamo and the compatibility between seismic and geomagnetic observations (e.g., the thickness of stratified layer) are still inconclusive (Buffett, 2014; Chulliat & Maus, 2014; Gubbins, 2007; Lesur et al., 2015). Additionally, the mechanism for the formation of stratification is also under debate. For example, high concentrations of light elements, including S, O, Si, C, and H, at the top of outer core could cause a stratification (e.g., Buffett & Seagle, 2010; Gubbins & Davies, 2013; Helffrich & Kaneshima, 2013; Fearn & Loper, 1981; Nakagawa, 2018;), but how these light elements change V_p is still under debate (Brodholt & Badro, 2017; Helffrich, 2012). In this study, we cannot easily distinguish between the EPOC and KHOMC models, but these two models do give different predictions of *SmKS-SKKS* differential arrival times. Thus, both gathering more observations, for example, *S3KS-SKKS* differential times at a distance $>160^\circ$, and considering other geophysical probes of the outer core, for example, normal mode observations, will be critical to better resolve the uppermost outer core's density and V_p , providing vital data to constrain the thermochemical status of the outer core.

6. Conclusions

We introduce an array-based iterative method to measure *SmKS-SKKS* ($m = 3, 4$, and 5) differential arrival times and use them to investigate the V_p in Earth's uppermost outer core. We validate this method by testing synthetic seismograms and apply this method to data at stations in Europe from 11 earthquakes in Fiji-Tonga, Vanuatu, New Britain, and Solomon Islands. Using the *SKKS* signal as a reference, *S3KS*, *S4KS*, and *S5KS* waveforms are successfully extracted and *S3KS-SKKS*, *S4KS-SKKS*, and *S5KS-SKKS* differential arrival times are measured by waveform CC. This iterative method not only gives us the measurements of differential arrival times but also allows us to assess measurement qualities based the CC coefficients and amplitude information. *SmKS-SKKS* differential arrival times are sensitive to V_p at the top of the outer core, but 3-D mantle structures could also affect the arrival times. We use the 3-D mantle model S40RTS and two different methods, ray theory and SEM synthetic seismograms, to estimate these anomalies for the frequency of 0.05–0.2 Hz. The results show that the arrival time anomalies due to 3-D mantle structure effects are large (e.g., >0.5 s) for some data, and sometimes there are big differences between the corrections calculated using ray theory and SEM synthetics. After corrections for 3-D mantle structure, we still see large positive *S3KS-SKKS*, *S4KS-SKKS*, and *S5KS-SKKS* differential arrival times, indicating a lower V_p than in PREM at the top of outer core. Our measurements are consistent with the predictions of KHOMC and EPOC models. EPOC has a homogeneous outer core while KHOMC contains a stratified layer at the top of outer core. Based on the data in this study, we cannot clearly distinguish the KHOMC and EPOC models, so more data, for example, *S3KS-SKKS* differential time at distances $>160^\circ$, will be necessary to help us distinguish between them.

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