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RESEARCH ARTICLE

Linking caregiving quality during infancy to brain activity in early childhood and later executive function

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Abstract

There is no relationship more vital than the one a child shares with their primary caregivers early in development. Yet many children worldwide are raised in settings that lack the warmth, connection, and stimulation provided by a responsive primary caregiver. In this study, we used data from the Bucharest Early Intervention Project (BEIP), a longitudinal study of institutionally-reared and family-reared children, to test how caregiving quality during infancy is associated with average EEG power over the first 3.5 years of life in alpha, beta, and theta frequency bands, and associations with later executive function (EF) at age 8 years. The sample comprised 189 children (129 institutionally-reared; 60 family-reared) who contributed data on observed caregiving quality during infancy (baseline; average age of 22 months), resting EEG power at baseline, 30, and 42 months, and performance-based data on a series of EF tasks at 8 years. Using Bayesian estimation, observed caregiving quality at baseline was marginally linked with higher average alpha and beta power, and lower theta power, from baseline to 42 months. In turn, higher average beta power and lower average theta power were marginally associated with higher EF at 8 years. In indirect effects models, higher caregiving quality at baseline was associated with higher EF at 8 years, with a marginal indirect effect through average theta power from baseline to 42 months. Variation in the quality of the early caregiving environment may be associated with later executive function, which is partially underpinned by individual differences in brain activity during early childhood.

KEYWORDS
brain activity, caregiving quality, EEG power, executive function

Research Highlights

- Examined associations between caregiving quality during infancy, brain activity during early childhood, and executive function during mid-childhood in sample of never-institutionalized and institutionally-reared children.
- Significant associations between higher quality caregiving during infancy and higher executive function during middle childhood.
- Marginal associations between caregiving quality during infancy and brain activity during early childhood.
- Marginal associations between brain activity during early childhood and executive function during mid-childhood.

1 | INTRODUCTION

1.1 | Caregiving and its consequences

Caregiving is one of the most fundamental and complex tasks performed by humans, safeguarding the survival of infants who depend entirely on their caregivers for nourishment, safety, and stimulation that ultimately lays the foundation for later socioemotional and cognitive development. In his seminal work, Bowlby described how the biobehavioral attachment system operates to maintain closeness between parents and their children, ensuring protection and maximizing the chance of survival (Bowlby, 1969/1982). However, caregivers serve a broader purpose in their children's development. They fulfill a dual role as a safe haven, offering comfort, and as a secure base, providing a launching point for infants to explore their surroundings (Ainsworth, 1973). These nurturing and supportive relationships serve as the basis for healthy child development. High-quality caregiving further facilitates development by modeling appropriate responses to stress and uncertainty, providing social, linguistic, and cognitive input, and scaffolding infants' understanding of societal values and cultural norms (Crockenberg & Leerkes, 2000; Rosenblum et al., 2009).

Despite wide cultural variation in the specific elements of parenting that contribute to healthy attachment relationships, it is generally the case that a child's relationship with their primary caregiver is critical to their psychosocial health and well-being (Ranson & Urichuk, 2008; Richter, 2004). This is true across a wide variety of caregiving contexts, including in societies where communal childrearing is the norm (Bornstein, 2012). However, due to factors such as orphanhood, abandonment, war, or sociopolitical turmoil, a minority of the world's children are raised in institutions where they do not have access to a stable caregiver who can reliably meet and sensitively respond to their needs. Studies of institutionally-reared children are crucial to developing a more comprehensive understanding of how the quality of caregiving impacts child development behaviorally and neurobiologically during the earliest years of life. Including these children is also important to more fully capture the range of caregiving quality that children are exposed to worldwide, and its implications for development. In the current study, we leverage data from an ongoing longitudinal study of institutionally-reared and family-reared children, which provides significant variation in the quality of caregiving received and could therefore improve prediction to later developmental outcomes.

Children raised in institutions face a number of threats to their development. In addition to disruption to their main attachment relationships (Doyle & Cicchetti, 2017), children raised in these settings experience more regimentation, exposure to peer aggression, less personalized and responsive care, and less psychosocial and cognitive stimulation that facilitates development in a number of domains. It is perhaps unsurprising, then, that children reared in institutions often have more mental health problems, more social and peer difficulties, greater risk of language and cognitive impairments, and altered neurobiological development (Van IJzendoorn et al., 2011).

1.2 | Caregiving quality and executive function

One domain of development that appears especially impacted by early deprivation in the context of institutional care is executive function (EF; Wade et al., 2019), a set of skills involved in problem solving, goal pursuit, and regulation of emotion and behaviour. EF is foundational to long-term adjustment. Recent meta-analytic work demonstrates that early EF predicts later school and employment success, social functioning, and mental and physical health across the lifespan (Robson et al., 2020). Importantly, EF is not only impacted by the extremes of poor quality caregiving as in the case of institutional care; rather, a significant body of literature suggests an association between caregiving quality and EF among family-reared children (Bernier et al., 2012; Bosquet Enlow et al., 2019; Cuevas et al., 2014). Caregiver factors such as scaffolding, stimulation, and sensitivity have demonstrated significant associations with child EF, especially during the highly-dependent early childhood years (Fay-Stammbach et al., 2014; Lucassen et al., 2015). Theoretical frameworks and correlational studies suggest that responsive caregivers may support their children's EF by acting as an external regulator, helping to scaffold children's responses to stimuli and modelling self-regulatory behavior during sensitive periods of development (Bernier et al., 2010; Greenough et al., 1987). Additionally, longitudinal research indicates that improvements in caregiving quality vis-à-vis entry into family care following institutional care buffers against the effect of later stressful life events on EF both behaviorally and neurologically (Wade et al., 2023). Longitudinal research conducted within typical home environments also suggests that early caregiving quality (measured at age 2 years) is related to later EF (measured at 4 years), highlighting the importance of early caregiving on the development of EF abilities (Hughes & Ensor, 2009). Furthermore, pathway analytic studies suggests that more emotionally supportive caregiving behaviour (e.g., caregiving with more positive affect) during infancy may be associated with improved EF, while more intrusive caregiving behaviour may have indirect effects on EF through changes in infants' neural activity (Swingler et al., 2018). Taken together, high-quality caregiving appears to act as both a promotive and protective factor for EF development among both institutionally-reared and family-reared children.

1.3 | Caregiving quality and brain activity

One promising pathway through which high-quality caregiving may be associated with later EF is through changes in brain function (Hane & Fox, 2006; Hane et al., 2010; Swingler et al., 2018). Specifically, both the serve-and-return interactions and protection from threatening experiences provided by responsive caregivers may shape neurobiological development in a way that fundamentally alters EF trajectories (Bourne et al., 2022). The electroencephalogram (EEG) provides a direct measure of brain activity and is especially useful to capture shifts in development across infancy and childhood (Bell & Cuevas, 2012; Buzsáki et al., 2012). Among non-institutionalized children, resting brain activity is typically characterized by an increase in higher-frequency band power (i.e., alpha and beta) and a decrease in lower-frequency band power (i.e., theta) from infancy to middle childhood (Marshall et al., 2002), as well as a shift in the peak power early in development for theta, and relatively later for alpha (Cellier et al., 2021). This is believed to reflect a process of cortical maturation during early childhood. Prior reports from the Bucharest Early Intervention Project (BEIP) showed that institutionally-reared children have higher resting EEG power in the theta band and lower power in the higher-frequency alpha and beta bands at baseline compared to never-institutionalized (i.e., family-reared) children (Marshall et al., 2004). This pattern has been interpreted as reflecting a maturational lag in functional brain development and has been associated with more attention and learning problems during childhood (Barry et al., 2003, 2009; Chabot et al., 2005; Mclaughlin et al., 2010). At 42 months, among institutionally-reared children, earlier (vs. later) placement into foster care was associated with more normalized resting brain activity (Marshall et al., 2008). Given that one of the salient differences between the institutionally-reared and family-reared children in the BEIP is early caregiving quality, this suggests that higher caregiving quality may be associated with indicators of more optimal brain development as indexed by resting EEG, perhaps especially in the first two years of life. Other intervention research has shown that preschool children whose parents participate in a program designed to enhance parental sensitivity show greater power in high-frequency beta bands (i.e., a reflection of cortical maturation) in middle childhood compared to those in the control group (Bick et al., 2019). Furthermore, maternal stress, a factor known to interfere with responsive parenting (de Cock et al., 2017; Enlow et al., 2019), has also been associated with this "lagged" profile of increased lower-frequency and decreased higher-frequency power early in life (Troller-Renfree et al., 2023). Thus, caregiving quality appears to be a key predictor of both concurrent and long-term brain development as captured via EEG.

1.4 | Brain activity and executive function

In addition to being responsive to the early caregiving environment, changes in brain activity have been hypothesized as a mechanism for cognitive development. Extant literature suggests that early signs of cortical maturation, as indicated by an increase in higher-frequency band power (i.e., alpha and beta), may underpin the development of higher order cognitive abilities such as EF later in childhood (Bhavnani et al., 2021; Fiske & Holmboe, 2019). For example, children who demonstrated an increase in resting alpha power from 10 months to 48 months were found to demonstrate greater inhibitory control, a critical component of EF, at 4 years (Whedon et al., 2020). This direct and positive association between increases in higher-frequency band power and EF is further demonstrated in cross-sectional research of preschoolers, where higher beta power has been linked to greater EF (Lo et al., 2013). Lastly, other theories posit that decreased theta power and increased beta power in children reflects a more well-developed goal-oriented attention system (Putman et al., 2014). Findings from Perone et al. (2018) support this relation, with a lower ratio of theta-to-beta power being linked with higher EF, even after controlling for age and verbal ability. Taken together, these findings suggest that resting EEG activity across the first few years of life may be predictive of EF development in later childhood.

1.5 | Current study

Extant literature clearly demonstrates the above-mentioned associations between caregiving and EF, caregiving and brain activity, and brain activity and EF. However, limited research has tested these pathways collectively in a single model with the primary outcome (i.e., EF) during middle childhood. Thus, the primary aim of the current study was to explore longitudinal associations between caregiving quality during infancy and brain activity over the first 3.5 years of life, and associations with later EF later at 8 years. While exploring later developmental outcomes in middle childhood does not explicitly permit casual or directional inferences, it helps to address a major limitation in this area of research (Bhavnani et al., 2021) – specifically, a dominant focus on preschool-aged children, which may be too narrow to adequately measure and interpret developmental changes of emerging cognitive abilities such as EF. Explorations of the association between early brain activity and EF during middle childhood are still scarce, with few studies addressing the gap of understanding age-related changes from early to middle childhood (Cai et al., 2021; Perone et al., 2018). Indeed, middle childhood may be an especially important period for EF development. EF is still developing significantly during this stage, with unique demands of this period requiring considerable use of EF skills (i.e., school and social contexts) alongside declining external support and scaffolding (Best et al., 2009; Engelhardt et al., 2019). Thus, the current study specifically tested whether the association between caregiving quality at baseline (average of 22 months) and EF at 8 years was mediated by mean resting EEG power in alpha, beta, and theta frequency bands, averaged from baseline to 42 months. The sample comprised both institutionally-reared and family-reared children, thus providing considerable variation on early caregiving quality. We hypothesized that higher caregiving quality at baseline would be associated with higher mean power in the alpha and beta bands, and lower power in the theta band, across time (baseline to 42 months). In turn, we expected higher average alpha and beta power, and lower theta power, to be associated with higher EF at 8 years, and for these measures of brain activity to partially mediate the association between caregiving quality during infancy on EF at 8 years.

2 | METHODS

2.1 | Participants and study design

Participants included children from the BEIP. Initially, 187 infants living in six institutions in Bucharest, Romania, were screened for inclusion. The institutions from which the children were recruited did not provide the social, cognitive, or linguistic input usually experienced during early childhood. The institutions were characterized by rigid schedules, high child-to-caregiver ratios, and rotating shifts of caregivers. Following the screening, 136 institutionally-reared children were enrolled into the study. Exclusionary criteria included frank neurological or genetic disorders, fetal alcohol syndrome, and micro- or macrocephaly. Also enrolled were 72 never-institutionalized community comparison children who were matched on age and sex to the institutionally-reared group. Following baseline assessments, the 136 institutionally-reared children were randomly assigned to either care-as-usual (i.e., remained in institutions; $n = 68$) or placement into a foster family through a network that was created and supported by the BEIP (foster care group, $n = 68$). All children were assessed at baseline (mean age of 22 months, range = 6–31 months), with follow-up assessments at 30, 42, and 54 months, at which point the trial officially ended and care of the children was taken over by local child protection agencies. Additional follow-up assessments took place at 8, 12, and 16 years, though the current study only uses data from the assessments at baseline (commenced April 2001; $N = 208$), 30 months (commenced May 2001; $N = 191$), 42 months (commenced April 2002; $N = 184$), and 8 years (commenced June 2007; $N = 215$).

In this study, 13 out of the original 208 children did not have baseline caregiving quality data, leaving 195 children who were considered for analysis. Two of these children had extreme values on one or more EEG variables, and four children were missing data on all other variables. After these exclusions, the final sample consisted of 189 children ($n = 129$ institutionally-reared; $n = 60$ never-institutionalized). Baseline descriptive statistics and simple group differences between institutionally-reared and never-institutionalized children are presented in Table 1.

2.2 | Measures

2.2.1 | Caregiving quality

The Observational Record of the Caregiving Environment (ORCE; NICHD Early Child Care Research Network, 1996) was used to rate caregiving quality, whether in the institutional or home environment. As described previously (Zeanah et al., 2005), the measure was adapted in two primary ways: First, rather than "live coding," research assistants videotaped children in interaction with their preferred caregiver for 1.5 h in either the institutional or home environments, thus facilitating a more objective assessment by independent coders. Caregivers were given no special instructions prior to the videotaping. Second, additional behaviors relevant to the experience of institutionally-reared children were qualitatively assessed, including marked dysregulation, stereotypical behavior, and communicative gesture. While both quantitative and qualitative items were scored, only the qualitative items were used in the present study. Qualitative coding occurred over the entire 1.5 h, which included a period in which quantitative scores were recorded (though these were not used here). With respect to coder training, coders were given a thorough orientation to both qualitative and quantitative items. They then completed 10 reliability tapes from either a local community project in New Orleans, or of recorded observations within the institutional settings during pilot testing. Differences between coders were resolved through discussion, and 40% of the tapes were then chosen for double-coding. The final Caregiving Quality score was obtained by averaging five qualitative scores—sensitivity, stimulation of development, positive regard for child, flat affect [reversed], and detachment [reversed])—each of which received a rating from 1 ("not at all characteristic") to 4 ("highly characteristic") and which were based on the coders' impression over the entire 1.5-h interaction. Scale reliability (Cronbach's alpha = 0.86) and inter-rater reliability ($M = 0.95$, range = 0.88–0.99) was previously shown to be excellent (Smyke et al., 2007). For the current sample ($n = 189$), scale reliability was also excellent (Cronbach's alpha = 0.87).

2.2.2 | EEG recording

Resting-state EEG was collected at baseline (20 months; $M_{age} = 20.40$ months; $SD = 7.20$ months), 30 months ($M_{age} = 30.84$ months;

TABLE 1 Sample characteristics for institutionally-reared and never-institutionalized children.

	Institutionally-reared ($n = 129$)		Never-institutionalized ($n = 60$)		Group difference	
	N (%)	*M* (*SD*)	*N* (%)	*M* (*SD*)	χ^2	*t*-test
Gender					0.33 (*n.s.*)	
Male	66 (51.2)		28 (46.7)			
Female	63 (48.8)		32 (53.3)			
Ethnicity					26.1***	
Romanian	71 (55.0)		54 (90.0)			
Roma	39 (30.3)		4 (6.7)			
Unknown	17 (13.2)		0 (0.0)			
Other	2 (1.6)		2 (3.3)			
Caregiving quality		2.16 (0.60)		2.81 (0.54)		7.12***
Alpha power		0.30 (0.03)		0.31 (0.03)		2.17*
Beta power		0.13 (0.02)		0.14 (0.02)		1.88†
Theta power		0.47 (0.04)		0.45 (0.04)		2.70**
Executive function		−0.29 (0.83)		0.50 (0.59)		6.37***

***$p < 0.001$.
**$p < 0.01$.
*$p < 0.05$.
†$p < 0.10$.

SD = 2.04 months), and 42 months (M_{age} = 42.36 months; SD = 1.44 months), while participants watched a bingo wheel. The resting-state EEG recording procedure has been previously reported (Marshall et al., 2004, 2008). The wheel had brightly colored balls that was spun by an experimenter for 90 s in 9 trials, each 10 s in length. Between trials, the experimenter stopped the spinning wheel and changed the balls in the wheel to maintain participants' attention for 10 s. EEG data analysis was performed on epochs in which the wheel was being spun (Marshall et al., 2004, 2008). EEG was collected using a lycra Electro-Cap (Electro-Cap International Inc., Eaton, OH) with sewn-in tin electrodes and the EEG Analysis System from the James Long Company (Caroga Lake, NY). Frontal (F3, F4, Fz), central (C3, C4, Cz), parietal (P3, P4, Pz), and occipital (O1, O2) electrodes were positioned according to the 10/20 System (Jasper, 1958). The vertex electrode (Cz) served as the reference, and an anterior midline electrode (AFz) served as the ground. Impedances of all sensors were kept <10 kΩ. EEG signals were digitized at a sampling rate of 512 Hz, and the amplifier filter settings were set at 0.1 Hz (high pass) and 100 Hz (low pass).

2.2.3 | EEG preprocessing and spectral analysis

EEG data were preprocessed using EEGLAB, an open-source analysis toolbox (Delorme & Makeig, 2004), as well as custom scripts written in MATLAB (version 2017a). EEG data were down sampled to 250 Hz and bandpass filtered (0.3 to 50 Hz). Continuous EEG data were then segmented into 2-s epochs with 1-second (50%) overlap. Epochs containing artifacts (e.g., eye blinks, movement) were identified using a voltage threshold of ±150 μV and rejected from analysis. Down-sampling, filtering, artifact rejection parameters were based on the parameters used in the Maryland Analysis of Developmental EEG (MADE) pipeline (Debnath, Buzzell, et al., 2020). Participants included in the final analysis had more than 20 s of artifact-free data. Epoched data were re-referenced to the average of two mastoid electrodes.

Using the cleaned data, a fast Fourier transform with a 2-s Hanning window and 50% overlap was performed. Absolute power was calculated for theta (baseline: 3–5 Hz; 30 months: 3–5 Hz; 42 months: 3–5 Hz;), alpha (baseline: 6–9 Hz; 30 months: 6–10 Hz; 42 months: 6–10 Hz), and beta (baseline:10–18 Hz; 30 months: 11–18 Hz; 42 months: 13–20 Hz) frequency bands. A natural log transformation was applied. The frequency ranges were based on work in this sample (Marshall et al., 2004, 2008) and studies showing slight differences in the frequency composition of the power spectra which changes across development (Marshall et al., 2002). We calculated and used relative power (i.e., absolute power of each frequency band divided by total absolute power across all frequency bands) averaged across electrodes (F3, F4, Fz, C3, C4, P3, P4, Pz, O1, O2) to produce a score across the whole scalp. We focused on relative power in the current longitudinal analyses because it has better test-retest consistency compared to absolute power. Also, relative power is more appropriate for examining developmental changes as it is less influenced by individual differences in impedance and skull thickness, which changes across development (Clarke et al., 2001).

2.2.4 | EEG data and scoring

The threshold for inclusion in the analysis was 20 s of artifact-free data. Participants who completed the EEG assessments included N = 185 (CAUG n = 67, FCG n = 66, NIG n = 52) at baseline, N = 161 (CAUG = 61, FCG = 61, NIG = 39) at 30 months, and *N* = 139 (CAUG = 53, FCG = 56, NIG = 30) at 42 months. For this specific analysis, there were *N* = 176 (CAUG = 64, FCG = 62, and NIG = 50) participants who contributed EEG data at baseline, *N* = 157 (CAUG = 59, FCG = 59, and NIG = 39) at 30 months, and *N* = 135 (CAUG = 51, FCG = 54, and NIG = 30) at 42 months.

The final EEG power variables used in the analyses were mean relative power in alpha, beta, and theta frequency bands that were averaged across baseline, 30 months, and 42 months, thus resulting in a single measure for each band capturing power over the first 3.5 years of life. The decision to average EEG power scores across the three measurement occasions was based on three preliminary findings. First, while it is possible to examine change in brain activity over time as a mediator of caregiving quality on EF, the pattern of change in the overall sample was, in fact, non-linear (see Figure S1); yet with only three waves of data collection, only a linear model can be reliably estimated. Second, the pattern of change in power over time was very similar for both institutionally-reared and never-institutionalized children for all three frequency bands (Figure S2). Specifically, the difference between these groups was in their overall level of brain activity, beginning at baseline (i.e., intercept in the latent growth model) and persisting through 42 months. There were no differences between the groups in the rate of change (i.e., the slope). Again, this rate of change was likely non-linear, and thus applying a linear model to the data may not accurately capture the shape of these developmental trajectories. Third, when examining rank-order stability in brain activity over time, we found that there were significant within-frequency band correlations over time (Table S1). Given the significant correlations between mean power measures over time within each frequency band, and given that the primary differences between the never-institutionalized and ever-institutionalized groups are in their initial power differences that are sustained over time—as opposed to their rate of change—we believe using the mean relative power for each frequency band averaged over time is justified and captures overall individual differences in power in this study (however, see below for Sensitivity Analysis using intercept and slope growth parameters as mediators).

2.2.5 | Executive function (EF)

EF was assessed using a well-validated behavioral measure, the Cambridge Neuropsychological Test Automated Battery (CANTAB). The CANTAB is a set of computerized tasks assessing memory and EF in a number of domains. Four subtests were administered: (1) *Delayed Match to Sample* (DMS, which measures attention and short-term visual memory); (2) *Stockings of Cambridge* (SOC, which measures spatial planning and problem-solving); (3) *Spatial Working Memory* (SWM, which measures the ability to update spatial information in memory); and (4) *Paired Associates Learning* (PAL, which measures visual-spatial memory and new learning). Tasks are described in detail on the CANTAB website (http://www.cantab.com). As described in Wade et al. (2020), single outcomes from each of the four tasks were selected and used

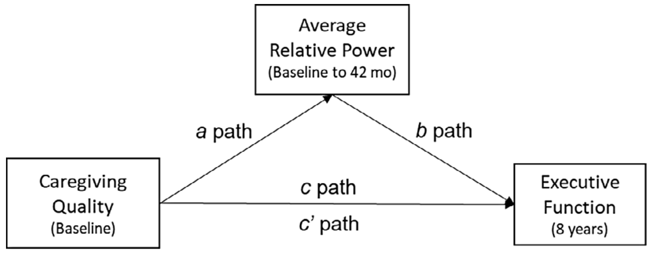

FIGURE 1 Structure of the path model linking caregiving quality at baseline to executive function at 8 years through average relative EEG power from baseline to 42 months. Three separate models were run for power in alpha, beta, and theta frequency bands, respectively.

as indicators of a global EF latent variable using confirmatory factor analysis. These indicators included DMS percent correct over all delays, SOC problems solved in the minimum number of moves, SWM total errors, and PAL mean errors to success. The EF latent factor was scaled such that higher DMS and SOC scores, and lower SWM and PAL scores, represented better memory, attention, and executive abilities. The model estimated EF at age 8, 12, and 16 years simultaneously, though for the current study we extracted and saved the factor scores at 8 years only.

2.3 | Statistical analysis

All children, both institutionally-reared and never-institutionalized, were included in the analysis. This provided wide variation in scores on the caregiving quality measure (range = 1.0–3.8). In descriptive analysis, we examined bivariate correlations between study variables, as well as group differences on sociodemographic and selected study variables. In the primary analysis, we tested three separate path models in which caregiving quality at baseline predicted EF at 8 years through average resting relative alpha (Model 1), beta (Model 2), or theta (Model 3) power from baseline to 42 months. We report total effects (*c* paths), direct effects (*c'* paths), and indirect effects (*a***b* paths), as well as the constituent paths comprising the indirect effect (i.e., caregiving quality predicting EEG power [*a* paths]; and EEG power predicting EF [*b* paths]). The general structure of these models is presented in Figure 1. For these models, path analysis in Mplus version 8.2 was used with Bayesian estimation with non-informative priors. In Mplus, Bayes uses full information from all observations, and is thus asymptotically equivalent to maximum likelihood estimation (Muthén & Asparouhov, 2012). Moderate amounts of missing data on either the EEG measures (*n* = 4; 2.1%) or EF scores (*n* = 43; 22.7%) were therefore handled using the Bayes estimator.

In using Bayesian estimation, interpretation is different than a standard two-tailed *p*-value in that 95% credibility intervals are interpreted as the interval that contains the population parameter with 95% probability (Hox et al., 2012). For example, a one-tailed Bayes *p*-value of 0.10 means that there is about a one-in-ten chance that the point estimate is actually zero or of the opposite sign, given the data (Hox et al., 2012). For the current study, we report unstandardized *B* coefficients with 95% credibility intervals (CIs), and interpret Bayes one-tailed p-values < 0.10 as providing marginal evidence for a given association. As outlined above, we hypothesized specific directions of the associations—namely, we hypothesized that higher caregiving quality would be associated with higher alpha and beta power, but lower theta power, and that higher alpha and beta, but lower theta, would be associated with higher EF. These directional hypotheses are reflected in the one-tailed Bayes p-value (Keysers et al., 2020), which is more diagnostic than a basic test for the existence of any effect, positive or negative (i.e., a two-tailed test; Marsman & Wagenmakers, 2017). However, we judge "significance" in the traditional way, where an effect is deemed significant if the 95% CI does not include zero. Model fit was evaluated and judged to be acceptable if the Posterior Predictive p-value was > 0.05 (with values closer to 0.50 representing excellent fit), and a 95% confidence interval for the difference between observed and replicated chi-square values that did not include zero.

In this study, we elected to include all participants (institutionally-reared and never-institutionalized) in the same models, without controlling for institutionalization group. This was because one of the most salient differences between these groups was, in fact, their level of caregiving quality (see Table 1). Thus, there would be significant shared variance between institutional group and caregiving quality variables, and controlling for the former would likely dramatically reduce, if not eliminate, the effect of the latter. Also, there are likely other significant differences between the institutionally-reared and never-institutionalized children that may further bear on brain activity and EF, yet our goal was to specifically examine individual differences in caregiving quality. Notwithstanding this point, as a sensitivity analysis, we: (1) tested whether any putative effects linking caregiving quality, brain activity, and EF were robust to controlling for group (i.e., never-institutionalized = 0 institutionally-reared = 1); (2) tested interactions between caregiving quality and institutional group on brain activity and EF to determine whether associations between caregiving quality and these outcomes varied as a function of group; (3) tested whether change in brain activity over time (i.e., the intercept and slope of EEG power in a latent growth model) mediated caregiving effects on EF; and (4) tested an alternative model with the ratio of theta-to-beta power (theta/beta ratio) as a mediator of caregiving effects on EF. We selected the theta/beta ratio since this is the theta ratio score that is most often linked with EF, attention, and ADHD in developmental science (Tan et al., 2023).

3 | RESULTS

3.1 | Descriptive analysis

Demographic and descriptive statistics between institutionally-reared and never-institutionalized children are presented in Table 1. Bivariate correlations and descriptive statistics for the entire sample are presented in Table 2. As expected and reported previously, there were differences between the institutionally-reared and never-institutionalized groups on most variables of interest, including significantly lower caregiving quality, significantly lower average alpha power, marginally lower average beta power, significantly higher average theta power, and significantly lower EF scores among the institutionally-reared children compared to never-institutionalized children. Across the entire sample, higher caregiving quality at baseline was marginally associated with lower average theta power and was significantly associated with higher EF. Higher average beta power, and lower average theta power, were also assigned with higher EF. The associations between average power from baseline to 42 months in alpha, beta, and theta bands with EF at 8 years are presented in Figure 2.

Model fit for each of the primary path models was excellent (see Table 3). The total effect of caregiving quality at baseline on EF at 8 years was significant in all three models with average alpha, beta, and theta power, respectively, as proposed mediators. In Model 1 (alpha power), there was marginal evidence for an association between higher caregiving quality and higher average alpha power (Bayes p-value = 0.07), but no evidence of an association between average alpha power and EF. Consequently, there was no evidence of an indirect effect of caregiving quality on EF through alpha power. In Model 2 (beta power), there was again marginal evidence for an association between higher caregiving quality and higher average beta power (Bayes p-value = 0.09). In this model there was also marginal evidence for an association between higher average beta power and higher EF at 8 years (Bayes p-value = 0.03). However, there was no evidence of an indirect effect of caregiving quality on EF through beta power. Finally, in Model 3 (theta power), there was marginal evidence for an association between higher caregiving quality and lower average theta power (Bayes p-value = 0.03). There was also relatively strong evidence for an association between lower average theta power and higher EF at 8 years (Bayes p-value = 0.01). In Model 3, there was marginal evidence for an indirect effect of caregiving quality on EF through theta power (Bayes p-value = 0.05). In all models, the direct effect of caregiving quality on EF was significant even whilst accounting for the effects of alpha, beta, and theta power, suggesting that EEG power is overall a relatively weak mediator of these associations.

3.2 | Sensitivity analysis

First, given that there is significant overlap between the quality of caregiving and group membership (i.e., being in the ever-institutionalized vs. never-institutionalized group), we re-ran the models with group as a binary covariate to determine whether the observed caregiving quality effects were significant over and above group membership. In doing so, the previously significant (or marginally significant) effect of caregiving quality on EEG power for all three frequency bands was reduced to non-significance, and this also eliminated the direct effect of caregiving quality on EF. After controlling for group, no significant indirect effects of caregiving quality on EF via EEG power were observed for any frequency band. In all of these models, group

TABLE 2 Bivariate correlations and descriptive statistics across the entire study sample.

	1.	2.	3.	4.	*M*	*SD*	*Range*
1. Caregiving quality	–				2.36	0.66	1.0–3.8
2. Alpha power	0.11	–			0.31	0.03	0.21–0.38
3. Beta power	0.10	−0.35***>	–		0.14	0.02	0.07–0.20
4. Theta power	−0.14†	−0.12†	−0.79***	–	0.47	0.04	0.35–0.59
5. Executive function	0.28**	0.13	0.18*	−0.23**	−0.01	0.85	−2.39–1.65

***$p < .001$.
**$p < .01$.
*$p < .05$.
†$p < .10$.

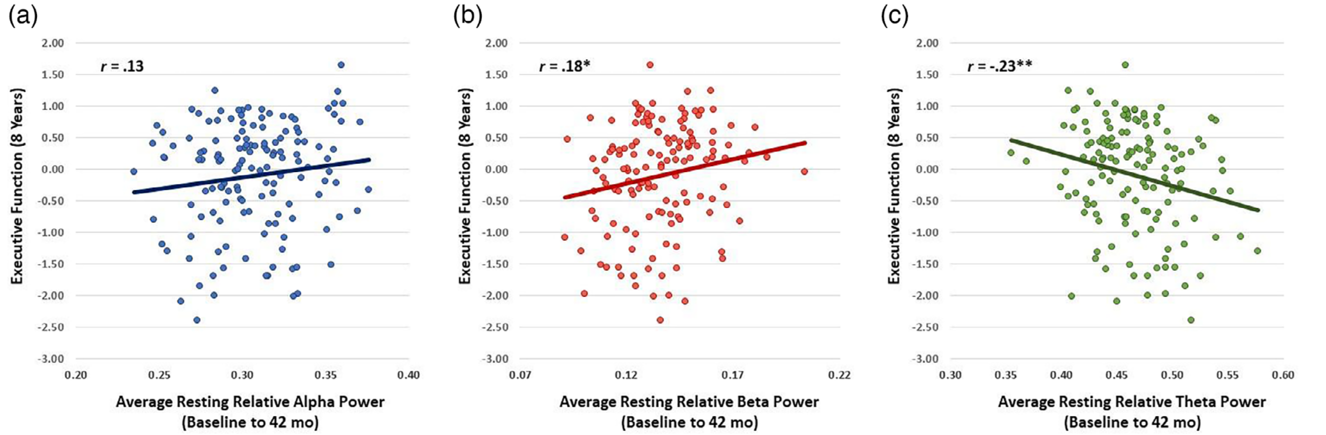

FIGURE 2 Associations between average relative alpha (a), beta (b), and theta (c) power from baseline to 42 months with executive function at 8 years. Significant correlations were observed for beta and theta power, but not alpha power, across the entire sample. All associations were in the expected direction. Coefficient *r* are the Pearson correlation coefficients.

was a highly significant predictor of caregiving quality, where being in the ever-institutionalized group was associated with almost a full standard deviation lower caregiving quality. Thus, caregiving quality does not exert a strong effect independent of group. This is perhaps unsurprising since one of the salient differences between the ever- and never-institutionalized groups is a significant difference in caregiving quality, in addition to a number of other differences not accounted for (and not measured) in the current study.

Second, we examined interactions between caregiving quality and group (never-institutionalized versus ever-institutionalized) in predicting EEG power in all three frequency bands, as well as EF. The results are presented in the Supplement (Table S2; Figure S3 and S4). Briefly, controlling for the main effect of group and caregiving quality, there was a significant interaction between these variables in predicting alpha power only, with simple slope analysis showing that the difference in alpha power between never- and ever-institutionalized groups was observed at lower but not higher levels of caregiving quality, suggesting that high-quality caregiving may partially offset the effect of institutional deprivation on early alpha power. However, this effect was marginal and should be interpreted cautiously.

Third, we tested whether the intercept and slope derived from a latent growth model mediated associations between caregiving quality and EF at 8 years. The results are presented in Figure S5. Briefly, neither the initial status (intercept) nor slope (rate of change) in mean relative power were associated with EF at 8 years for any frequency band, though there was a marginal association between higher initial theta at baseline and lower EF at 8 years. Moreover, caregiving quality was associated with higher alpha power at baseline and lower theta power at baseline, and was associated with the slope of alpha power, with higher caregiving associated with less growth (i.e., a less steep slope) over time. This finding may be explained by the significant inverse association between the intercept and slope, where those with higher initial alpha power experienced less of an increase in alpha power over time. Importantly, these results included slope and intercept parameters derived from a linear model (since there were only three waves of data), yet the pattern of change over time may be non-linear (Figures S1 and S2). These results should therefore be interpreted cautiously.

Finally, we tested an alternative model with the ratio of mean relative theta power to mean relative beta power (theta/beta ratio), averaged from baseline to 42 months, as a mediator of the association between caregiving quality at baseline and EF at 8 years. The results are presented in the Supplement (Figure S6), and are largely consistent with the primary results involving theta power alone in which marginal evidence for mediation was observed, with the same overall pattern of associations linking caregiving quality, brain activity, and EF.

TABLE 3 Total, direct, and indirect effects linking caregiving quality to executive function through brain activity.

	Model 1: Alpha power as mediator		Model 2: Beta power as mediator		Model 3: Theta power as mediator	
	Unstandardized (Standardized) effect	95% CI (unstandardized effect)	*Unstandardized (Standardized) effect*	95% CI (unstandardized effect)	*Unstandardized (Standardized) effect*	95% CI (unstandardized effect)
Total effect (*c* path)	**0.35***** (0.26)	0.13, 0.56	**0.35***** (0.27)	0.14, 0.56	**0.34**** (0.26)	0.13, 0.56
Direct effect (*c′* path)	**0.34**** (0.25)	0.12, 0.55	**0.33**** (0.25)	0.12, 0.54	**0.31**** (0.23)	0.10, 0.52
Caregiving quality→EEG (*a* path)	0.01† (0.11)	0.00, 0.01	0.003† (0.10)	0.00, 0.01	−0.01* (−0.14)	−0.02, 0.00
EEG→EF (*b* path)	2.65 (0.09)	−2.02, 7.20	6.43* (0.15)	−0.54, 13.3	**−3.98*** (−0.19)	−7.63, −0.47
Indirect effect (*a*b* path)	0.01 (0.01)	−0.01, 0.06	0.02 (0.01)	−0.01, 0.07	0.03* (0.02)	0.00, 0.09
Model Fit						
PPP	0.495		0.493		0.494	
95% CI	−10.55, 10.73		−10.52, 10.89		−10.53, 10.83	

Notes: Effects flagged at ***$p < 0.001$. **$p < 0.01$. *$p < 0.05$. †$p < 0.10$ reflect the one-tailed Bayes *p*-value for the given directional hypothesis. Bolded coefficients are significant at $p < 0.05$ (two-tailed). See Figure 1 for structural model and path labels. 95% CI—95% credibility interval from Bayesian regression analysis.

Abbreviations: EF, executive function as assessed using the Cambridge Neuropsychological Test Automated Battery; PPP, posterior predictive *p*-value.

4 | DISCUSSION

Warm, sensitive, and responsive caregiving is essential to healthy development, yet the specific pathways through which caregiving quality impacts child outcomes are not fully understood. Considering the plasticity of the brain early in life (Berardi et al., 2015), it is conceivable that nurturing interactions with caregivers help to shape early neurobiological development in a way that promotes cognitive function during childhood. Further, theoretical frameworks highlight how caregiver-child interactions are central in determining the pace of neurobiological development (Roubinov et al., 2021). Specifically, these models suggest that, in response to deprived early caregiving environments, there may be a developmental *deceleration* across neurocognitive domains (e.g., brain activity, EF) in order to adapt to unmet physiological, physical, and interpersonal needs. The results of the current study are consistent with these models, showing that caregiving quality in infancy was marginally associated with average EEG power in three canonical frequency bands (alpha, beta, and theta) during the first 3.5 years of life, with lower caregiving quality associated with a pattern that has been interpreted to reflect slower cortical maturation (lower high-frequency power and higher low-frequency power). In turn, lower beta power and higher theta power were marginally associated with lower EF at age 8 years. Marginal evidence of mediation was observed for theta power only, though it is important to note that the effects reported in this study were generally small. The results together suggest that caregiving quality during infancy is associated with EF abilities through middle childhood (Koşkulu-Sancar et al., 2023; Werchan et al., 2023), and that individual differences in brain activity may partially underlie these associations.

Caregiving quality was operationalized across the dimensions of sensitivity, stimulation, positive regard for child, positive affect, and attachment. Considering these dimensions align with the concept of "serve-and-return" in which responsiveness to a child's cues and appropriate stimulation during infancy scaffolds cognitive development (see National Scientific Council on the Developing Child, 2004; Shonkoff & Bales, 2011), it was predicted that higher quality caregiving in infancy would be associated with better EF in middle childhood. Results supported this hypothesis, as children exposed to higher caregiving quality during the first two years of life had better developed EF abilities at 8 years. These findings were consistent in all three models that included alpha, beta, and theta power as potential mediators. The association between caregiving quality during infancy and EF at 8 years was the most robust finding in the current study and was supported whether we relied on the directional Bayes one-tailed *p*-value (Keysers et al., 2020) or more traditional two-tailed hypothesis testing. By including children with histories of institutional care and those raised in their biological families, we were able to capture a wide range in children's early caregiving experiences, thus improving prediction to later outcomes. However, sensitivity analysis showed that these associations did not persist after controlling for institutionalization history, which likely reflects, in part, significant differences in caregiving quality as a function of this history. Overall, our results are consistent with decades of work in developmental science that have underscored the association between high quality early care and children's long-term cognitive outcomes.

Although this study focused on caregiving quality during infancy, we should not assume that this is the only, or even the most important, period during which caregiving matters for development. Our measure of caregiving quality at baseline was significantly correlated with ORCE-assessed caregiving quality at 30 and 42 months of age, and with rater-assessed caregiving quality at 54 months and 8 years (the age of EF assessment in the current study). Thus, there is some stability in caregiving quality over time. We have previously shown that significant differences in rater-assessed caregiving quality persist between study groups at 8, 12, and 16 years and that, over and above group status, rater-assessed caregiving quality at these ages is associated with

EF, with stronger associations between caregiving quality and EF over time (Colich et al., 2021). Thus, although we see an association between observed caregiving quality during infancy and later EF in the current study, testing how either stability or change in caregiving quality is associated with stability or change in EF, and whether these associations are mediated by brain activity over time, is an important question for future research.

The second aim of this study was to examine associations between caregiving quality and brain activity (i.e., resting EEG power), and brain activity as a potential mediator of EF. Relying on the directional Bayes one-tailed *p*-value, there was marginal evidence that higher caregiving quality was associated with higher average alpha and beta power, and lower theta, from baseline to 42 months. This increase in alpha and beta power, with a respective decrease in theta power, is believed to be a general metric of cortical maturation (Marshall et al., 2002). Although there was a clear signal in the data when relying on the Bayes one-tailed test which was consistent with study hypotheses, results were not significant when using the traditional $p < 0.05$ threshold in two-tailed testing. Thus, the effect of caregiving quality on EEG power were overall marginal, though it should be noted that the relatively small sample may have also contributed to difficulties reaching traditional levels of significance. These results suggest that, while caregiving may be associated with resting brain activity, there may be other potential biological and psychosocial factors that contribute to changes in resting brain activity during early childhood, including socioeconomic factors, maternal stress, the home language environment, and genetic factors (e.g., Jensen et al., 2021; Orekhova et al., 2003; Pierce et al., 2021).

Resting brain activity was then investigated as a potential mediating mechanism linking caregiving quality during infancy to EF in middle childhood. It was hypothesized that the neurobiological profile of increased average alpha and beta power, and decreased theta power, averaged from baseline to 42 months, would be linked to greater EF at 8 years and partially explain the relation between caregiving quality and later EF. There was some evidence that this EEG profile was directionally associated with EF at 8 years. Specifically, directional associations between higher beta power and lower theta power with higher EF were observed when relying on the directional Bayes one-tailed *p*-value. No evidence of an association between alpha power and EF was observed. When relying on the two-tailed test, only theta power was significantly associated with EF. Thus, the effects of resting EEG power on EF were modest and appear to be strongest for theta power. These results diverge somewhat from literature linking increased alpha and beta power to components of EF (Lo et al., 2013; Whedon et al., 2020). This may be explained by the components of EF captured in the current study, which mainly indexed memory and attentional components of EF as opposed to the inhibitory dimensions of some prior studies (Lo et al., 2013; Whedon et al., 2020). It is also important to note that our findings slightly differ from studies conducted with children in developmentally-expectable ecobiological environments (i.e., family reared children; Lo et al., 2013; Whedon et al., 2020), whereas almost two-thirds of our sample were raised in institutional settings. Thus, the current study's unique sample may also contribute to these divergent findings. Moreover, the results of our sensitivity analysis were consistent with those of Perone et al. (2018), who showed that a lower theta/beta ratio was associated with better performance on a tablet-based EF task indexing working memory and cognitive flexibility among 3–9 year old children. Thus, it is possible that different domains of EF show distinct associations with certain EEG parameters early in development. However, the high association between alpha and beta power, and the similar direction of association with EF for both frequency bands, suggest that the lack of association between alpha power and later EF may have been due to low power. Replication in larger samples is therefore strongly encouraged.

Finally, with respect to the indirect effects of caregiving quality on EF via EEG power, there was evidence from the Bayes one-tailed *p*-value that higher caregiving quality at baseline was associated with higher EF at 8 years through lower average theta power from baseline to 42 months. No evidence of mediation was found when relying on the traditional two-tailed *p*-value. These results suggest that theta power may be a mechanism linking caregiving quality to EF in middle childhood. However, considering the relative weakness of this effect, and the fact that the direct association between caregiving quality and later EF continued to be significant after accounting for EEG power, there are likely additional unmeasured biopsychosocial pathways connecting early caregiving to later EF. For example, it is possible that the early caregiving environment impacts EF more immediately and strongly via bottom-up response systems that govern stress, emotion, and memory processes. Specifically, recent literature suggests that warm and responsive caregiving during early sensitive periods may affect corticolimbic circuitry (Gee & Cohodes, 2021) which, in animal models, has been shown to impact self-regulatory behavior related to EF (see Blair, 2010). Further, existing literature from the BEIP data demonstrates a causal link between the caregiving environment and physiological reactivity to social stress (Mclaughlin et al., 2015), which has been linked to lower EF-related processes in some studies (e.g., Raffington et al., 2018). Early adversity has been associated with blunted cortisol reactivity to social stress which is, in turn, related to lower EF in late childhood (Conradt et al., 2014). To the extent that EEG power is not fully indexing these other biological pathways, the current results suggest that the mechanisms linking caregiving quality during infancy to EF during middle childhood are likely complex and multidimensional, with EEG power being one potential pathway. Future research that considers the intersection of stress system responsiveness, brain activity, and EF in the context of both early and later caregiving quality is warranted based on these preliminary results.

The strengths of this study include its longitudinal design that allows for exploration of the mediating pathways from early caregiving quality to EF development in middle childhood. Further, this study has analytical strengths, as it employed Bayesian estimation which has emerged as a preferable approach (compared to frequentist methods) when dealing with complex path models and directional hypotheses (Hox et al., 2012; Keysers et al., 2020; Muthén & Asparouhov, 2012). Additionally, this study employed diverse methodology to measure the constructs of interest, including objective coding of caregiving quality, high-fidelity measurement of EEG with recent advances in preprocessing, and direct performance-based assessment of EF. This diversification of methods and measures helps guard against biases that emerge from reliance on, for example, parent reports of both caregiving and behavior. Finally, the current study includes children raised in both family and institutional settings. This inclusion is both statistically advantageous by allowing for wide variation on the predictor variable of interest (caregiving quality) and significant in terms of real-world implications, as it enables us to capture the various levels of caregiving quality experienced by children globally, thereby enhancing our understanding of its impact on their development. Notably, children who experienced institutional care were shown, on average, to have lower caregiving quality than family-reared children. Controlling for institutional care history eliminated most of the observed effects, suggesting that the effect of caregiving quality on brain activity and EF was not independent of placement history in this study. Nevertheless, there was significant variation in caregiving quality within both groups (institutional and family reared), and it therefore seems appropriate to have focused on individual differences in caregiving quality as opposed to group differences.

There are also several limitations of this study. First, although we demonstrated several effects of interest, they were generally small and of marginal significance. While not a limitation per se, this may reflect the fact that the study was underpowered to detect certain effects, especially mediated effects (Fritz & MacKinnon, 2007). Second, while directional hypotheses were often supported by relying on the Bayes one tailed *p*-values, the results should not be taken to represent strong evidence for the effect of caregiving quality on early EEG power, nor of EEG power on EF. Given that intervention effects on EEG power in the BEIP have not been demonstrated until later in childhood (Debnath, Buzzell, et al., 2020; Vanderwert et al., 2016), it is possible that the benefits of high-quality early caregiving on brain activity are only beginning to emerge early in life, with stronger effects during late childhood or adolescence. This would underscore the possibility of a so-called "sleeper effect" of early caregiving quality on brain development, with possible knock-on effects for later cognition. Indeed, we have shown such sleeper effects previously in the BEIP (Mukerji et al., 2021; Wade et al., 2018), and have demonstrated that caregiving quality during adolescence is associated with a range of outcomes above and beyond the early caregiving environment (Colich et al., 2021). Thus, assessing whether the effects of caregiving quality on brain activity and EF becomes stronger over the course of development is an important topic for future research. Relatedly, testing whether earlier versus later caregiving quality is more strongly linked to EF over the course of childhood and adolescence is necessary to determine whether there are so-called timing effects that may reflect periods of increased sensitivity to the caregiving environment. Third, the current study examined whole-brain relative power as a mechanism linking early deprivation to later EF, though it is possible that these effects manifest more strongly in specific regions of the brain (e.g., frontal regions). In other words, early deprivation may be more focally associated with EEG power for specific frequency bands in specific regions of the brain. This would, in turn, increase the precision of effects and potentially reveal a more reliable set of mechanistic findings. However, given that most findings in the BEIP involving group effects on power, and power effects on cognition, have focused on whole-brain relative power (Debnath, Tang, et al., 2020; Tan et al., 2023; Vanderwert et al., 2016; Wade et al., 2020), and without a reliable set of prior findings on which to base region-specific hypotheses, we limited the current analysis to examine whole-brain power. Fourth, while potentially limited in its power to detect certain effects, this study also underscores the fact that additional mechanisms not presently measured are likely to explain the relatively robust relation between caregiving quality during infancy and EF during middle childhood. Finally, this study only includes children living in Bucharest, Romania, with two-thirds of the sample having spent some portion of their lives in profound conditions of deprivation within institutions. While the importance of early caregiving quality has been demonstrated across regions and cultural contexts, both the ethnic homogeneity of this sample and the unique early experience of many children who participated may impact generalizability.

5 | CONCLUSION

This study emphasizes the importance of caregiving quality during the first 2 years of life on resting brain activity over the first 3.5 years and EF during middle childhood. Moreover, it expands knowledge on the mechanisms through which caregiver quality may be associated with later EF, with individual differences in brain activity—in particular, resting theta power and the theta/beta ratio—serving as one potential pathway to later EF. Importantly, these pathways were not robust to controlling for institutional care history, perhaps, in part, because of the significant overlap between caregiving quality and group status. This underscores the need to differentiate between caregiving quality and other institutional characteristics in future work. Mapping the complex biopsychosocial pathways through which caregiving quality scaffolds EF over time is important to detect problems before they emerge using non-invasive techniques and technologies so that early intervention can be offered to those at the greatest risk. Before this can occur, a clearer delineation of the role of EEG power and its reliability in these pathways is needed, as is a more comprehensive charting of the factors that translate high quality care into positive developmental outcomes.

AUTHOR CONTRIBUTIONS

Conceptualization: Mark Wade, Charles A. Nelson, Charles H. Zeanah, Nathan A. Fox; Methodology: Mark Wade, Charles A. Nelson, Nathan A. Fox, Charles H. Zeanah; Analysis & Visualization: Mark Wade; Funding Acquisition: Charles A. Nelson, Nathan A. Fox, Charles H. Zeanah; Project Administration: Charles A. Nelson, Charles H. Zeanah, Nathan A. Fox; Writing: Mark Wade, Victoria Parker; Critical Review and Editing: Alva Tang, Charles A. Nelson, Nathan A. Fox, Charles H. Zeanah.

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CONFLICT OF INTEREST STATEMENT

NAF has received grant support from the National Institute of Mental Health (NIMH), the Eunice Kennedy Shriver National Institute of Child Health and Human Development (NICHD), the National Science Foundation (NSF), the National Institutes of Health Environmental influences on Child Health Outcomes (NIH ECHO) consortium, the Russell Sage Foundation, and the Lumos Foundation. He has received royalties from Guilford Press and Harvard University Press. He has received honoraria for lectures to professional audiences. CHZ has received grant support from the NIMH, the Palix Foundation, the Irving Harris Foundation, the Substance Abuse and Mental Health Services Administration (SAMHSA), the Lumos Foundation, and the Inter-American Development Bank. He has received royalties from Guilford Press and Harvard University Press. He has received consulting fees from the National Society for the Prevention of Cruelty to Children. He has received honoraria for lectures to professional audiences. CAN has received grant support from the National Institutes of Health (NIH), the Jacobs Foundation, the John D. and Catherine T. MacArthur Foundation, the James S. McDonnel Foundation, the Binder Family Foundation, the Lumos Foundation, the Bill and Melinda Gates Foundation, and Harvard University. He has received royalties from the Massachusetts Institute of Technology and Harvard University Press. He has received honoraria for lectures to professional audiences and legal consulting fees. MW, VP, and AT report no competing or potential conflicts of interest.

DATA AVAILABILITY STATEMENT

The data and analytic code necessary to reproduce the analyses, and the materials necessary to attempt to replicate the findings, are not publicly accessible. However, both the data and the code are available from the first author upon reasonable request. The analyses presented here were not preregistered.

ETHICS APPROVAL STATEMENT

The original study was approved by the institutional review boards of the three principal investigators [CAN, CHZ, NAF] and by the local Commissions on Child Protection in Bucharest (Humphreys et al., 2017).

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SUPPORTING INFORMATION

Additional supporting information can be found online in the Supporting Information section at the end of this article.

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