Deterministic replica-exchange method without pseudo random numbers for simulations of complex systems

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We propose a replica-exchange method (REM) which does not use pseudo random numbers. For this purpose, we first give a conditional probability for Gibbs sampling replica-exchange method (GSREM) based on the heat bath method. In GSREM, replica exchange is performed by conditional probability based on the weight of states using pseudo random numbers. From the conditional probability, we propose a new method called deterministic replica-exchange method (DETREM) that produces thermal equilibrium distribution based on a differential equation instead of using pseudo random numbers. This method satisfies the detailed balance condition using a conditional probability of Gibbs heat bath method and thus results converge to the Boltzmann distribution. We confirmed that the equivalent results were obtained by REM and DETREM with two-dimensional Ising model. DETREM can avoid problems of choice of seeds in pseudo random numbers for parallel computing of REM.

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INTRODUCTION

The enhancement of sampling during Monte Carlo (MC) and molecular dynamics (MD) simulations is very important for complex systems. Replica-exchange method (REM) (or parallel tempering) is one of the most popular ways to improve sampling efficiency [1–4] (for reviews, see, e.g., Refs.[5, 6]). To realize a thermal equilibrium distribution, REM uses Metropolis criterion with pseudo random numbers. However, random numbers sometimes give inaccurate results [7]. For example, in MD, bad random numbers could cause synchronization of trajectories, which decreases the effectiveness of sampling [8]. On the other hand, generation of high quality random numbers is often difficult and does not assure good simulation results [9]. REM is suited for parallel computing, while most of pseudo random number generators decrease the scalability in parallelization [10]. Hence, the complementary method producing the same results without pseudo random numbers is meaningful.

Recently, Suzuki et al. proposed a method to produce a thermal equilibrium state without using random numbers for spin models by a differential equation based on the conditional probability of Gibbs sampling heat bath method, which is referred to as chaotic Boltzmann machines [11, 12]. The differential equation controls spin states at each site and the staying time of each spin state is proportional to the weights of the thermal equilibrium distribution. They reproduced the results of a conventional MC method in some spin systems.

We here generalize this method to REM. We first have to extend the conditional probability for replica exchange not based on Metropolis criterion but on a Gibbs sampling heat bath method. The heat bath formalism has already been given in Ref. [13], we refer to this method as Gibbs sampling replica-exchange method (GSREM). (A similar approach based on global balance condition [14] was also developed [15].) We then introduce a differential equation for replica exchange to modify GSREM. This method is referred to as the deterministic replica-exchange method (DETREM). We then tested the effectiveness of DETREM by comparing the results of simulation of 2-dimensional Ising model with those by the conventional REM.

The organization of this paper is as follows. In Section 2, the theory for the new method and conventional REM is presented. In Section 3, we give the results of DETREM together with REM. The final section is devoted to conclusions.
METHODS

We first briefly review the conventional REM. We prepare $M$ non-interacting replicas at $M$ different temperatures. Let the label $i (=1, \cdots, M)$ stand for the replica index and label $m (=1, \cdots, M)$ for the temperature index. Here, $i$ and $m$ are related by the permutation functions by

$$\begin{aligned}
i &= i(m) \equiv f(m), \\
m &= m(i) \equiv f^{-1}(i),
\end{aligned} \tag{1}$$

where $f(m)$ is a permutation function of $m$ and $f^{-1}(i)$ is the inverse. We represent the state of the entire system of $M$ replicas by $X = \{x_{m(1)}, \cdots, x_{m(M)}\}$, where $x_{m}^i = \{q^i_m, p^i_m\}$ are the set of coordinates $q^i$ and momenta $p^i$ of particles in replica $i$ (at temperature $T_m$). The probability weight factor for state $X$ is given by a product of Boltzmann factors:

$$W_{\text{REM}}(X) = \prod_{i=1}^{M} \exp[-\beta_m(x^i)H(q^i_m, p^i_m)], \tag{2}$$

where $\beta_m = 1/k_B T_m$ is the inverse temperature and $H(q, p)$ is the Hamiltonian of the system. We consider exchanging a pair of replicas $i$ and $j$ corresponding to temperatures $T_m$ and $T_n$, respectively:

$$X = \{\cdots, x_n^i, \cdots, x_n^j, \cdots\} \rightarrow X' = \{\cdots, x_n^j', \cdots, x_n^i', \cdots\}, \tag{3}$$

where $x_n^i = \{q^i_n, p^i_n\}$, $x_n^j = \{q^j_n, p^j_n\}$, and $p^i_n = \sqrt{\frac{k_B T_m}{m}} p^i_n, p^j_n = \sqrt{\frac{k_B T_m}{m}} p^j_n$. The exchange of replicas introduces a new permutation function $f'$:

$$\begin{aligned}
i &= f(m) \rightarrow j = f'(m), \\
j &= f(n) \rightarrow i = f'(n). \tag{4}
\end{aligned}$$

We remark that this process is equivalent to exchanging a pair of temperatures $T_m$ and $T_n$ for the corresponding replicas $i$ and $j$.

Here, the transition probability $\omega(X \rightarrow X')$ of Metropolis criterion is given by

$$\omega(X \rightarrow X') = \min\left(1, \frac{W_{\text{REM}}(X')}{W_{\text{REM}}(X)}\right) = \min(1, \exp(-\Delta)), \tag{5}$$

where

$$\Delta = \Delta_{m,n} = (\beta_n - \beta_m)(E(q^i) - E(q^j)). \tag{6}$$

REM is performed by repeating the following two steps:

1. We perform a conventional MD or MC simulation of replica $i (=1, \cdots, M)$ at temperature $T_m$ ($m = 1, \cdots, M$) simultaneously and independently for short steps.

2. Selected pairs of replicas are exchanged based on the above Metropolis criterion in Eqs. (5) and (6). A pseudo random number is used to judge the criterion.

Without loss of generality we can assume $T_1 < T_2 < \cdots < T_M$. Note that in Step 2 we usually exchange only pairs of replicas corresponding to neighboring temperatures, because the acceptance probability for replica exchange decreases exponentially with the difference of the two inverse temperatures and potential energy terms because of Eq. (6). This replica exchange can be written as

$$X = \{\cdots, x_m^i, \cdots, x_m^j, \cdots\} \rightarrow X' = \{\cdots, x_m^j, \cdots, x_m^i, \cdots\}, \tag{7}$$

where in Eq. (5) $\Delta$ is now given by

$$\Delta_m = (\beta_{m+1} - \beta_m)(E(q^i) - E(q^j)). \tag{8}$$
The REM method makes a random walk in temperature space during the simulation. The canonical ensemble is reconstructed by the multiple-histogram reweighting technique, or weighted histogram analysis method (WHAM) [16, 17].

We next present GSREM [18]. For REM with heat bath method, the conditional probability assigned for new states for any replica-exchange is given by

$$\omega(S \mid x^{[1]}, x^{[2]}, \ldots, x^{[M]}) = \frac{W(x^{[1]}_{m(1)}, x^{[2]}_{m(2)}, \ldots, x^{[M]}_{m(M)})}{\sum_{S' \in S_M} W(x^{[1]}_{m'(1)}, x^{[2]}_{m'(2)}, \ldots, x^{[M]}_{m'(M)})},$$

(9)

where $S \equiv \{m(1), m(2), \ldots, m(M)\}$ is a permutation of temperature indices and $S_M$ is all possible permutations. In GSREM, the above procedure for the conventional REM is performed, where Step 2 for the GSREM is performed based on Eq. (9). Here, in Step 2, the conditional probability of a temperature set based on Eq. (9) is calculated, and this assigns weights between 0 and 1 for for exchanged states and a no-exchange state. Finally, after a pseudo random number is generated, the state corresponding to the random number with the assigned region is selected.

When we consider exchange of only one pair of replicas to reduce the set $S_M$, Eq. (9) turns into

$$\omega(x^{[i]}_{m(i)}, x^{[j]}_{m(j)} \mid x^{[k \neq m(i), m(j)]}_{m(k)}) = \frac{W(x^{[i]}_{m(i)}, x^{[j]}_{m(j)})}{\sum_{i' = 1}^{M-1} \sum_{j' > i'} W(x^{[i']}_{m(i')}, x^{[j']}_{m(j')})},$$

(10)

As in the conventional REM, we usually consider the neighboring temperature exchange in Eq. (7). The conditional probability $\omega(x^{[i']}_{m(i')}, x^{[j']}_{m(j')} \mid x^{[k \neq i(m), j(m+1)]}_{m(k)})$, in which the new state selects the temperature exchanged state of replicas $i$ and $j$ with $T_{m+1}$ and $T_m$ from the no-exchange state of replicas $i$ and $j$ with temperatures $T_m$ and $T_{m+1}$, is given by

$$\omega(x^{[i']}_{m(i')}, x^{[j']}_{m(j')} \mid x^{[k \neq i(m), j(m+1)]}_{m(k)}) = \frac{W(x^{[i']}_{m(i')}, x^{[j']}_{m(j')} \mid x^{[k \neq i(m), j(m+1)]}_{m(k)})}{W(x^{[i']}_{m(i')}, x^{[j']}_{m(j')} \mid x^{[k \neq i(m), j(m+1)]}_{m(k)}) + W(x^{[i]}_{m(i')}, x^{[j]}_{m(j')} \mid x^{[k \neq i(m), j(m+1)]}_{m(k)})}$$

$$= \frac{1}{1 + \frac{W(x^{[i]}_{m(i')}, x^{[j]}_{m(j')} \mid x^{[k \neq i(m), j(m+1)]}_{m(k)})}{W(x^{[i']}_{m(i')}, x^{[j']}_{m(j')} \mid x^{[k \neq i(m), j(m+1)]}_{m(k)})}},$$

(11)

(12)

For the Boltzmann distribution, this equation in Eq. (11) can be rewritten as

$$\omega(x^{[i']}_{m(i')}, x^{[j']}_{m(j')} \mid x^{[k \neq i(m), j(m+1)]}_{m(k)}) = \frac{1}{1 + \exp(\Delta_m)},$$

(13)

where $\Delta_m$ is given by Eq. (8). This is the Gibbs sampling replica-exchange method when an equilibrium state is produced by this conditional probability with pseudo random numbers.

We next propose DETREM. At first, we introduce an internal state $y$ to assign a permutation of temperature state based on Eq. (9). It satisfies the following differential equation:

$$\frac{dy}{dt} = \frac{W(x^{[1]}_{m(1)}, x^{[2]}_{m(2)}, \ldots, x^{[M]}_{m(M)})}{\sum_{S' \in S_M} W(x^{[1]}_{m'(1)}, x^{[2]}_{m'(2)}, \ldots, x^{[M]}_{m'(M)})},$$

(14)

where $t$ is a virtual time, $y \in [1, N_{S_M} + 1]$, and $N_{S_M}$ is the total number of elements in the permutation set $S_M$. This DETREM is performed just like GSREM, where in Step 2 the evaluation of the conditional probability in Eq. (9) is replaced by solving the differential equation in Eq. (14). DETREM gives the same probability as in Eq. (9). When $y$ reaches $m = \lfloor y \rfloor$, the permutation of temperature corresponding to the integer in temperature permutations is chosen. Here, the floor function $\lfloor y \rfloor$ is the integral part of $y$ defined by $\lfloor y \rfloor = m \leftrightarrow m \leq y < m + 1$. In general, $N_{S_M}$ is so large that using Eq. (14) is not practical.
We thus introduce an internal state $y_{m,n}$ for exchange of a pair of temperatures $T_m$ and $T_n$ that correspond to replicas $i$ and $j$, respectively, with the time integration based on Eq. (10), which gives

$$
\frac{dy_{m,n}}{dt} = \frac{W(x_n^{[1]}, x_n^{[b]})}{W(x_n^{[1]}, x_n^{[b]}) + W(x_n^{[1]}, x_n^{[b]})} = \frac{1}{1 + \exp(\Delta_{m,n})},
$$

(16)

where $y_{m,n} \in [0,1]$ and $\Delta_{m,n}$ is given by Eq. (9).

As in the conventional REM, we can only use the internal states $y_m \in \{-1,1\}$ for a pair of neighboring temperatures $(T_m, T_{m+1})$, where the number of internal states is $M-1$ with the following pairs: $y_1 = (T_1, T_2), y_2 = (T_2, T_3), \ldots, y_{M-1} = (T_{M-1}, T_M)$. Hence, the differential equation based on Eq. (19) is given by

$$
\frac{dy_m}{dt} = \sigma_m \frac{1}{1 + \exp(\Delta_m)},
$$

(17)

where $\Delta_m$ is given by Eq. (8) and the signature $\sigma_m$ of the pair of $(T_m, T_{m+1})$ changes to 1 or $-1$ to control the signature of the change of $y_m$ which monotonically increases or decrease.

Compared to REM, the difference of the algorithms is in Step 2. In Step 2 of DETREM, instead of evaluating the

$\sigma_m = 1$ or $-1$ for new $y_m$ after exchanges are made. We remark that DETREM is performed just like GSREM, where in Step 2 the evaluation of the conditional probability in Eq. (13) by pseudo random numbers is replaced by solving the differential equation in Eq. (17).

Expectation values of physical quantities are given as functions of temperatures by WHAM[16–18]. Namely, the density of states $n(E)$ and dimensionless Helmholtz free energy are obtained by solving the following equations self-consistently:

$$
n(E) = \frac{\sum_{m=1}^{M} N_m(E)}{\sum_{m=1}^{M} n_m e^{f_m - \beta_m E}},
$$

(19)

and

$$
e^{-f_m} = \sum_{E} n(E) e^{-\beta_m E},
$$

(20)

where $N_m(E)$ and $n_m$ are the energy histogram and the total number of samples obtained at temperature $T_m$, respectively. After we obtained $f_m$ at each temperature, the expectation value of a physical quantity $A$ at any
temperature $T$ is given by

$$< A >_T = \frac{1}{\sum_{m=1}^{M} \sum_{x_m} A(x_m) \frac{1}{\sum_{l=1}^{M} n_l \exp \left( -f_l \beta_l E(x_m) \right)}} \exp \left( -\beta E(x_m) \right) \exp \left( -\beta E(x_m) \right),$$

(21)

where $x_m$ are the set of coordinates at temperature $T_m$ obtained from the trajectories of the simulation.

We remark that simulated tempering (ST)\cite{13, 20} corresponding to GSREM and DETREM can also be formulated. The weight factor for ST is given by

$$W_{m}^{ST}(x) = \exp \left[ -\beta_m E(x) + f_m \right],$$

(22)

where $f_m$ are the dimensionless Helmholtz free energy at temperature $T_m \ (m=1, \cdots, M)$. In Step 1, we perform a canonical MC or MD simulation at temperature $T_m$ for short steps. In Step 2, temperature is updated to a new value $T_n$ with the following conditional probability:

$$\omega(T_n \mid x) = \frac{W_{n}^{ST}(x)}{\sum_{m=1}^{M} W_{m}^{ST}(x) \sum_{m=1}^{M} \exp \left[ -\beta_m E(x) + f_m \right]} \propto \exp \left[ -(\beta_n - \beta_m) E(x) + (f_n - f_0) \right],$$

(23)

where we have introduced an arbitrary reference temperature $T_0$ to give the normalization. This is the transition probability for the Gibbs sampling simulated tempering (GSST) (this formulation was first given in Ref.\cite{13}).

From above, we can derive a differential equation for deterministic simulated tempering (DETST). We introduce an internal state $y$

$$\frac{dy}{dt} = \exp \left[ -(\beta_n - \beta_0) E(x) + (f_n - f_0) \right],$$

(25)

where $y \in \{1, \cdots, M+1\}$ and $m = \lfloor y \rfloor$.

In another implementation, the conditional probability from the current temperature $T_m$ to $T_n$ is given by

$$\omega(T_n \mid x, T_m) = \frac{W_{n}^{ST}(x)}{W_{n}^{ST}(x) + W_{m}^{ST}(x)} \cdot \frac{1}{1 + \Delta_{m,n}^{ST}}.$$

(26)

Thus, we introduce an internal state $y_{m,n}$ integrated by

$$\frac{dy_{m,n}}{dt} = \frac{W_{n}^{ST}(x)}{W_{n}^{ST}(x) + W_{m}^{ST}(x)} \cdot \frac{1}{1 + \Delta_{m,n}^{ST}},$$

(27)

where $y_{m,n} \in \{0,1\}$, $m, n \in \{1, 2, \cdots, M\}$, and

$$\Delta_{m,n}^{ST} = (\beta_n - \beta_m) E(x) - (f_n - f_m).$$

(29)

When we consider temperature change to neighboring values\cite{21}, the conditional probability from temperature $T_m$ into $T_{m+1}$ or $T_{m-1}$ is given by

$$\omega(T_{m \pm 1} \mid T_m, x) = \frac{W_{m \pm 1}^{ST}(x)}{W_{m \pm 1}^{ST}(x) + W_{m}^{ST}(x)} \cdot \frac{1}{1 + \Delta_{m \pm 1}^{ST}} \cdot \frac{1}{1 + \Delta_{m}^{ST}},$$

(30)

(31)
where $\Delta_{\pm}^{ST}$ are defined by

$$\Delta_{\pm}^{ST} = (\beta_{m\pm 1} - \beta_{m})E(x) - (f_{m\pm 1} - f_{m}). \quad (32)$$

Hence, the internal states and differential equations are given by

$$\frac{dy_m^+}{dt} = \frac{1}{1 + \Delta_{+}^{ST}}, \quad (33)$$

$$\frac{dy_m^-}{dt} = \frac{1}{1 + \Delta_{-}^{ST}}, \quad (34)$$

where $y_m = y_{m,m+1} \quad (m = 1, \cdots, M - 1)$. When the system stays at temperature $T_m$, only $y_m^\pm$ are updated, and other $y_m^{\pm, n \neq m}$ are not updated.

Simulation conditions

In order to test the effectiveness of the present methods, we studied the 2-dimensional Ising model. The lattice size $L$ in square lattice was 128. The system size $N$ is equal to $L^2$. For REM, replica-exchange attempt was made for every 1 MC step. 1 MC step was defined by the number of degrees of freedom. The total number of MC steps was 100,000,000. To integrate Eq. (17), we used the fourth-order Runge-Kutta method, which is equivalent to Eq. (18), with virtual time step $dt = 1$. The total number of replicas was 40 and the temperatures were 1.50, 1.55, 1.60, 1.65, 1.70, 1.75, 1.80, 1.85, 1.90, 1.94, 1.98, 2.01, 2.04, 2.07, 2.10, 2.13, 2.16, 2.19, 2.22, 2.25, 2.28, 2.31, 2.34, 2.358, 2.368, 2.38, 2.40, 2.42, 2.44, 2.47, 2.51, 2.57, 2.63, 2.69, 2.75, 2.82, 2.90, 3.00, 3.10, and 3.15. Boltzmann constant $k_B$ and the coupling constant $J$ were set to 1. Thus, $\beta = 1/k_B T = 1/T = \beta^*$, and the (potential) energy is given by

$$E(s) = -\sum_{<i,j>} s_i s_j, \quad (35)$$

where $s_i = \pm 1$, and the summation is taken over all the nearest-neighbor pairs in the square lattice. The canonical distribution is given by

$$W(s) = \frac{1}{Z}\exp(-\beta^* E(s)), \quad (36)$$

where $Z$ is the partition function. In the DETREM simulation, all $y_m$ were updated simultaneously. The multiple exchanges of temperatures at a replica were prohibited. Namely, if the neighboring internal states $y_m$ and $y_{m+1}$ satisfy the exchange condition ($y_m \geq 1$ or $y_m \leq -1$), only one state, e.g., $y_m$, was updated and only the pair $(T_m, T_{m+1})$ was exchanged, while $y_{m+1}$ was not updated.

RESULTS

Figs. 2(a) and 2(b) show the time series of temperature change in one of the replicas (Replica 1) as a function of MC steps from the conventional REM and the DETREM simulation, respectively. They show similar behaviors with respect to random walks in temperature space. We see that all replicas take the minimum temperature many times during both simulations. Other replicas perform random walks similarly. Figs. 3(a) and 3(b) show the time series of replica index at the minimum temperature of 1.5 during the REM and DETREM simulations. This shows that all replicas experienced the minimum temperature many times during the simulation. Table 4 lists the maximum number of tunneling events per replica, which is the number of times where the simulation visits from the lowest temperature through the highest temperature and back to the lowest temperature. These data show that the two methods have nearly the same number of tunneling counts during the simulations. All these results imply that REM and DETREM are equally efficient in sampling.

We next examine physical quantities obtained from the DETREM simulation and compare them to those from the REM simulation. Fig. 4 shows the canonical energy distributions at 40 temperatures as functions of energy obtained from the REM and DETREM simulations. We see that the distributions have enough overlaps in pairs of the neighboring distributions. This ensures that the number of replicas is sufficient. The agreement between two
TABLE I. The number of maximum tunneling count (TC) per replica of whole replicas during simulations

|       | TC REM | DETREM |
|-------|--------|--------|
| Max   | 191    | 196    |
| Mean ± SD | 173.3 ± 9.5 | 177.95 ± 8.8 |

SD means standard deviation with respect to replicas.

methods implies that DETREM method produced the Boltzmann distributions at each temperature simulation just like REM did.

We next confirm the second-order phase transitions at the critical temperature of $T_c \sim 2.269$ in both methods. Fig. 5(a) and Fig. 5(b) show the total energy density $\epsilon$ as a function of $T$ during the REM simulation and the DETREM simulation, respectively, where $\epsilon$ is defined by

$$\epsilon = \frac{E}{N}. \quad (37)$$

Fig. 6(a) and Fig. 6(b) show the specific heat $C$ as a function of $T$ from the REM simulation and the DETREM simulation, respectively, where $C$ is defined by

$$C = \frac{1}{kT^2N}(<E^2> - <E>^2). \quad (38)$$

Fig. 7(a) and Fig. 7(b) show magnetization $M$ as a function of $T$ from the REM simulation and the DETREM simulation, respectively, where $M$ is defined by

$$M = \frac{\sum_{i=1}^{N} s_i}{N}. \quad (39)$$

Fig. 8(a) and Fig. 8(b) show susceptibility $\chi$ as a function of temperature during the REM simulation and the DETREM simulation, respectively, where $\chi$ is defined by

$$\chi = \frac{N}{kT}(<M^2> - <M>^2). \quad (40)$$

All these physical quantities confirm that the DETREM simulation reproduced the results of the REM simulation in the phase transitions near the critical temperature $T_c$.

CONCLUSIONS

In this work, we proposed a deterministic replica-exchange method, which enables replicas to exchange their temperatures keeping their thermal equilibrium without using pseudo random numbers. We reproduced the results of REM by DETREM. DETREM can give thermal equilibrium quantities without using random numbers. This fact may be useful for parallel computing because bad seeds of generating random numbers cause less efficiency of simulations since parallel computing program code often uses same seeds during simulations.

This new method can easily be applied the transformation of conditional probability to other replica-exchange method variants. In a future work, we will introduce the multidimensional DETREM for generalized potential function including Hamiltonian replica-exchange method\textsuperscript{[22, 23]}

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FIG. 1. Schematic sketch of DETREM. After parallel conventional MC or MD simulations at $M (=6$, here) different temperatures for short steps, all internal states $y_m$ are updated. The temperature pairs $(T_m, T_{m+1})$, or corresponding replica pairs, are exchanged when $y_m \geq 1$ or $\leq -1$. The cycle is repeated until the end of simulation. T1, T2, T3, T4, T5, and T6 are temperatures. R1, R2, R3, R4, R5, and R6 are replicas.

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FIG. 2. Time series of temperature change in one of the replicas (Replica 1) for (a) REM and (b) DETREM simulations.

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FIG. 3. Time series of replica at temperature 1.5 for (a) REM and (b) DETREM simulations.

FIG. 4. Canonical probability distributions of energy at each temperature from (a) REM and (b) DETREM simulations. The distributions correspond to all the 40 temperatures (from the lowest temperature (1.5), the left-most, to the highest temperature (3.15), the right-most).
FIG. 5. Average energy density as a function of temperature obtained by WHAM from the (a) REM and (b) DETREM simulations.

FIG. 6. Specific heat as a function of temperature obtained by WHAM from the (a) REM and (b) DETREM simulations.
FIG. 7. Magnetization as a function of temperature obtained by WHAM from the (a) REM and (b) DETREM simulations.

FIG. 8. Susceptibility as a function of temperature obtained by WHAM from the (a) REM and (b) DETREM simulations.