Figure S1. The main procedure of GA.

Figure S2. An example of the BMS approach.
LIBS dataset (p variables, n samples) (6144 \times 240 in this study)

Set the parameters of VCPA, including the number of sampling runs, k; the EDF runs, N; the mean number of sampled variables of all runs, Alpha; the ratio of best models of k sub-models, Ratio_better.

While i < N runs of EDF

NO

Generate a binary matrix M (k \times p) and randomly permutation by column.

YES

Calculate the RMSECV of each subset in each row of M.

Sort all the k RMSECV. Compute the frequency of variables from the best ratio subsets.

Compute the left number of variables using EDF. Retain the variables based on the frequency.

After N runs, there are L variables left for the further optimization.

IGA

IRIV

Figure S3. Flow chart of VCPA.