

1 **Title Page**

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3 Manuscript title:

4 **Optimization of Air Monitoring Networks Using Chemical Transport Model and Search**

5 **Algorithm**

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29 **Abstract**

30 Air monitoring network design is a critical issue because monitoring stations should be  
31 allocated properly so that they adequately represent the concentrations in the domain of interest.  
32 Although the optimization methods using observations from existing monitoring networks are  
33 often applied to a network with a considerable number of stations, they are difficult to be applied  
34 to a sparse network or a network under development: there are too few observations to define an  
35 optimization criterion and the high number of potential monitor location combinations cannot be  
36 tested exhaustively. This paper develops a hybrid of genetic algorithm and simulated annealing to  
37 combine their power to search a big space and to find local optima. The hybrid algorithm as well  
38 as the two single algorithms are applied to optimize an air monitoring network of PM<sub>2.5</sub>, NO<sub>2</sub> and  
39 O<sub>3</sub> respectively, by minimization of the mean kriging variance derived from simulated values of a  
40 chemical transport model instead of observations. The hybrid algorithm performs best among the  
41 algorithms: kriging variance is on average about 4% better than for GA and variability between  
42 trials is less than 30% compared to SA. The optimized networks for the three pollutants are  
43 similar and maps interpolated from the simulated values at these locations are close to the  
44 original simulations (RMSE below 9% relative to the range of the field). This also holds for  
45 hourly and daily values although the networks are optimized for annual values. It is demonstrated

46 that the method using the hybrid algorithm and the model simulated values for the calculation of  
47 the mean kriging variance is of benefit to the optimization of air monitoring networks.

48

49 **Keywords:** PM<sub>2.5</sub>; NO<sub>2</sub>; O<sub>3</sub>; Genetic algorithm; Simulated annealing; Japan

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## 52 **1. Introduction**

53 Air monitoring networks have been developed in various areas in the world for environmental,  
54 epidemiological, policy evaluation and/or emission surveillance purposes by national or local  
55 governments. These networks should be allocated properly so that they adequately represent the  
56 concentrations in the domain of interest to accomplish the purposes of the network. This issue is  
57 often referred to as a network design problem and has been widely discussed (e.g., Brus et al.,  
58 2007; Wu et al., 2011). The network design problem usually aims at minimization of a design  
59 criterion that may be based on observations or other information about the field of interest: to  
60 achieve this aim, subsets of potential monitor locations are selected by an algorithm.

61 A design criterion is often defined with the notion of entropy where a set of locations which  
62 maximize the entropy at the monitored sites is searched for (e.g., Zidek et al., 2000; Fuentes et al.,  
63 2007). Another popular criterion is defined with a geostatistical estimation method which is  
64 called kriging, where the theoretical interpolation error averaged over the region of interest, i.e.  
65 the mean kriging variance is minimized (e.g., Baume et al., 2011; Wu et al., 2011). When the  
66 mean kriging variance is used as a criterion, observations obtained from existing monitoring  
67 network are often used to construct a variogram for the calculation of the mean kriging variance.  
68 In these cases, it is assumed that the network represents the spatial distribution sufficiently, thus  
69 the network of interest is relatively dense where the efficiency of the network is focused, i.e.

70 reduction of stations. Therefore, this method is difficult to apply to a sparse network or a network  
71 under development that insufficiently represents the spatial distribution of the pollutant of interest.

72 In the field of air quality study, the chemical transport model (CTM), that simulates physical  
73 and chemical processes including emission, advection, photochemical reactions and deposition,  
74 has been extensively used at various ranges of spatial and temporal scale, not only to obtain a  
75 spatial distribution, but also to establish an effective strategy for the control of the concentrations  
76 of air pollutants (e.g., Emmons et al., 2011; Chatani et al., 2014). Thus, simulated concentrations  
77 from CTM with sufficiently high spatial resolution can be an alternative to observations to derive  
78 a variogram to compute the mean kriging variance as a design criterion. Simulations have also  
79 been used for sampling optimization by Kumral and Ozer (2013) in mine planning.

80 Once the criterion is defined, the network design problem can be treated as a combinatorial  
81 optimization problem. When a network is small enough, complete enumeration of all possible  
82 combinations is possible. For a large network, however, this will run into a combinatorial  
83 explosion. To deal with this difficulty, search algorithms have been applied to the optimization of  
84 large networks. For instance, Ruis-Cardenas et al. (2010) applied genetic algorithm (GA) for an  
85 O<sub>3</sub> monitoring network with several hundred stations in the United States. Wu et al. (2011)  
86 applied simulated annealing (SA) to the optimization of an O<sub>3</sub> monitoring network over France.  
87 Given that the search space is huge when using simulated fields, superior ability both for a global

88 and a local search is required for a search algorithm to be applied. GA is able to search in a large  
89 space, but is often not able to find the local optimal solution (Ruiz et. al., 2010). On the other  
90 hand, SA is able to find locally optimal solutions, but is often trapped in regions far from the  
91 global optimum (Ruiz et. al., 2010). Araki et al. (2015) developed a hybrid of GA and SA (HGS)  
92 and successfully applied it to the optimization of a PM<sub>2.5</sub> monitoring network using simulated  
93 values obtained from CTM for the computation of the mean kriging variance. However, the  
94 performance of HGS was not compared to those of other optimization algorithms in their study,  
95 thus the advantages of HGS have not been demonstrated. In addition, HGS was applied only for a  
96 PM<sub>2.5</sub> monitoring network, and the possibility of application to other pollutants, that might have  
97 different spatial distribution features, was not examined.

98 In this paper, CTM is used to generate spatial distributions of air pollutant concentrations to  
99 derive variograms for the computation of the mean kriging variance as a design criterion, and  
100 each of the algorithm including HGS, GA and SA is applied to PM<sub>2.5</sub>, NO<sub>2</sub> and O<sub>3</sub> in the Kinki  
101 region of Japan respectively, repeating each setting 30 times to capture random effects. The  
102 performances of the algorithms are compared against each other in terms of the quality of the  
103 solutions such as the mean and the standard deviation of the mean kriging variance of the  
104 respective trials. Fields have been interpolated by ordinary kriging using the CTM simulated  
105 values at the selected sites in the optimized networks, and the errors between the interpolated and

106 simulated fields are computed. The difference between the algorithms for each of the pollutants is  
107 discussed and the capability and the applicability of HGS are evaluated.

108

## 109 **2 Methodology**

### 110 **2.1 Chemical Transport Model**

111 The chemical transport model used in this study is the Community Multiscale Air Quality model  
112 (CMAQ) (Byun and Ching, 1999) version 5.0.1 which was driven with the Weather Research and  
113 Forecasting model (WRF) (Skamarock et al., 2009) version 3.5.1. Meteorological fields were  
114 produced using WRF configured with the same physics options as those used by Shimadera et al.  
115 (2014); also emission data for the air quality simulations was produced in a similar way. The  
116 other settings involved in the simulations and CMAQ configurations are detailed in Shimadera et  
117 al. (2015).

118 The WRF/CMAQ model was run from April 2010 to March 2011 (Japanese fiscal year 2010)  
119 with an initial spin-up period of 22-31 in March 2011. The horizontal domains consisted of three  
120 domains: domain 1 covering a wide area of Northeast Asia, domain 2 covering the main land of  
121 Japan, and domain 3 covering the area where the optimization algorithms are tested, which is  
122 shown as colored area in Figure 1. The horizontal resolution is 4 km and the number of grids is  
123  $68 \times 72$  for domain 3. The annual and daily values used for the network optimization are



124 computed by averaging the hourly CTM outputs over the corresponding time periods. This  
125 simulation is identical to that used in Araki et al. (2015).

126 The performance of the model is detailed in Shimadera et al. (2015) and summarized as follows:  
127 the statistical measures obtained from the comparison between observed and simulated daily  
128 concentrations indicate that the model simulates the temporal and spatial variation patterns of  
129  $PM_{2.5}$ ,  $NO_2$  and  $O_3$  well with the Pearson's correlation coefficient being 0.76, 0.82, and 0.77 for  
130  $PM_{2.5}$ ,  $NO_2$  and  $O_3$  respectively. The scatter plots of the simulated and observed values of annual  
131 means for  $PM_{2.5}$  and  $NO_2$ , and annual means of daily maximum 8-hr mean concentrations for  $O_3$ ,  
132 which are used to derive variograms for the computation of the mean kriging variance, are  
133 presented in Figure 2 with root mean squared error (RMSE) and  $R^2$  between the observed and  
134 simulated values. The number of observations for  $PM_{2.5}$ ,  $NO_2$  and  $O_3$  is 8, 219 and 188  
135 respectively. The reason for the limited number of observations for  $PM_{2.5}$  is because the  $PM_{2.5}$   
136 network in Japan started to be developed since 2009, a year before the target year of this  
137 simulation. The concentrations of  $O_3$  lie in a relatively narrow range, which results in low  $R^2$   
138 value for  $O_3$ . However, RMSE of  $O_3$  is approximately 10% of the mean values of  $O_3$ . Therefore,  
139 these simulated concentrations have sufficient quality to derive variograms for the computation of  
140 the mean kriging variance as the optimization criterion. The spatial distributions of  $PM_{2.5}$ ,  $NO_2$   
141 and  $O_3$  are given in Figure 1. Both  $PM_{2.5}$  and  $NO_2$  show highest concentrations in densely

142 populated areas, where  $\text{NO}_2$  is more concentrated to the agglomeration of Osaka. The higher and  
143 lower concentration areas of  $\text{PM}_{2.5}$  are generally distributed evenly. On the other hand, higher  
144 concentrations of  $\text{NO}_2$  are found in limited areas where megacities are located, while the lower  
145 areas are widely distributed. The spatial distribution of  $\text{O}_3$  is generally the reverse of that of  $\text{NO}_2$ .  
146 This is because the  $\text{O}_3$  concentrations are affected by titration with  $\text{NO}_x$ , and higher  $\text{NO}_x$   
147 concentrations might cause lower  $\text{O}_3$  concentrations in urban areas with relatively large  
148 anthropogenic emissions. The histograms of the simulated values in the candidate areas i.e. in the  
149 colored area in Figure 1 are given in Figure 3, which reflect the characteristics of the distribution  
150 of each pollutant:  $\text{PM}_{2.5}$  concentrations are approximately normally-distributed, the distribution  
151 of  $\text{NO}_2$  concentrations is skewed to zero and that of  $\text{O}_3$  is relatively skewed to higher values.

## 152 **2.2 Design criterion**

153 The optimization criterion is minimizing the mean kriging variance, which has been used as a  
154 design criterion in many previous studies (e.g., Van Groenigen, 2000; Brus et al., 2007; Wu et al.,  
155 2010; Wu et al., 2011; Baume et al., 2011). A definition of a design criterion is an expression of a  
156 purpose of a monitoring network. When minimization of the mean kriging variance is used as a  
157 criterion, the purpose of the network can be seen to grasp the spatial distribution of a pollutant in  
158 the whole region of interest well.

159 The simulated concentrations are transformed to a natural logarithmic scale before analysis, and  
160 back transformed after analysis so that the predicted concentrations are positive, which is not  
161 always the case without transformation (Beelen et al., 2009). The concentrations simulated by  
162 CTM are treated as point values at the grid cell centroids and an empirical variogram is computed  
163 from all the candidate points. A theoretical variogram is fitted to it with spherical model by  
164 weighted least-squares fitting. The derived variograms for the three pollutants are given in Figure  
165 4. Although the ranges of the three variograms are slightly different, the variograms are generally  
166 similar, with nugget very close to zero.

167 The fitness value for a selected set of points is the mean kriging variance: kriging variance is  
168 computed in all points of the grid in the region of interest from the variograms and the selected  
169 set of points and averaged over the area; to avoid overestimation of the kriging variance at the  
170 boundaries, fifteen equally-spaced points at the margins of the region of interest (see Figure 1)  
171 are in addition regarded as selected.

### 172 **2.3 Optimization algorithms**

173 The optimal combination of points is searched for by the optimization algorithms. Each  
174 network configuration is given by a binary string, where each element represents a candidate  
175 location, with ones at gauged sites and zeros at ungauged sites. A subset of locations is selected  
176 from the candidate locations according to this binary string.

### 177 **2.3.1 Genetic algorithms**

178 The basic scheme of GA algorithm in this paper is similar to the standard GA which consists of  
179 population, selection, crossover and mutation. The initial population is randomly generated and  
180 the population size is 100. In the crossover operation, gauged sites in both parents are gauged in  
181 the offspring, and ungauged sites in both parents are ungauged in the offspring. Half of the sites  
182 that are gauged in one of the parents and not gauged in the other are randomly conversed. The  
183 probability of crossover occurrence is set to 0.9. In the mutation operation, 5% of gauged sites are  
184 randomly selected and swapped with the same number of randomly selected ungauged sites with  
185 a mutation probability of 0.2. The solutions with the best 5% fitness, called elite, remain in the  
186 next generation, while others are rejected in order to keep the population size unchanged. These  
187 operations stop when the total number of generations reaches 500. The algorithm also stops when  
188 the number of generations without any improvement in the fitness reaches 40.

### 189 **2.3.2 Simulated annealing**

190 SA is an iterative search algorithm which starts with a randomly selected initial solution and the  
191 fitness value is computed. A new potential solution is created by exchanging a randomly selected  
192 gauged site for a randomly selected ungauged site in the pre-defined search window of the  
193 selected gauged site. The fitness value is then computed for the new potential solution and  
194 compared to that of the current solution. The new potential solution is accepted when the fitness

195 value improves. Even when the fitness value degrades, the new potential solution is accepted with  
196 a certain probability, which is given by

$$197 \quad p = \exp\left(\frac{-\Delta f}{T}\right), \quad (1)$$

198 where  $\Delta f$  is the amount of degradation of the fitness value and  $T$  is a control parameter (Brus et al.,  
199 2007).  $T$  is gradually decreased with iterations by

$$200 \quad T_{k+1} = \alpha T_k, \quad (2)$$

201 where  $\alpha$  is 0.9 and  $k$  is a number of iterations. The initial  $T$  (i.e.,  $T_1$ ) is set so that the average  $p$  in  
202 the equation (1) equals to 0.8 (Brus et al., 2007).

203 The size of the search window is fixed in the first 500 iterations at 18 km (approximately equals  
204 to  $7 \times 7$  cells window). Then it is decreased after 500 iterations down to 12 km ( $5 \times 5$  window),  
205 and 6 km ( $3 \times 3$  window) from 1,000 iterations and onward. The total number of iterations for  
206 each run is set to 50,000. The algorithm also stops when the number of iterations with no  
207 progress in the fitness value reaches 4000.

### 208 **2.3.3 Hybrid algorithm**

209 HGS algorithm is identical to that applied in Araki et al. (2015) and summarized as follows. The  
210 basic scheme of HGS is similar to GA. The initial population of the size of 100 is randomly  
211 generated and GA is applied for 75 iterations. After 75th generation, SA is applied to each of the  
212 15% best solutions for 30 iterations with the fixed search window size of  $7 \times 7$ . The rest 85%

213 solutions are kept unchanged and combined with the solutions obtained from SA. GA is then  
214 applied to this set of solutions i.e. population for 1 iteration, and the next generation is obtained.  
215 Again, SA is applied to the best 15% solutions for 30 iterations in the generation and the  
216 solutions are combined to the rest 85% solutions. The next generation is obtained by the  
217 application of 1 iteration of GA to the set of solutions. The parameters of HGS in the first 75  
218 generations are identical to GA algorithm described above. After 75 generations, the crossover  
219 and mutation probability is set to 0.3 and 0 respectively. After the 100th generation, the search  
220 window size is reduced to  $5 \times 5$ . This window size is again decreased to  $3 \times 3$  after 125th  
221 generation and is keep unchanged. The total number of iterations is set to 155, but the algorithm  
222 also stops when the number of generations with no progress in the fitness value reaches 8 after  
223 75th generation. The flowchart of HGS is presented in Figure 5.

#### 224 **2.4 Application of the algorithms**

225 The algorithms are tested in the area that includes the Kinki region in Japan ( $134.2^{\circ}\text{E} -$   
226  $136.5^{\circ}\text{E}$ ,  $33.4^{\circ}\text{N} - 35.8^{\circ}\text{N}$ ) that consists of six prefectures, and its contiguous areas, which equals  
227 to the colored area in Figure 1. Only land areas are considered for the potential locations of the  
228 monitoring stations. The number of the candidate locations is 1984. Megacities such as Osaka,  
229 Kyoto and Kobe are located in this region. The number of monitoring stations for  $\text{NO}_2$  and  $\text{O}_3$  in  
230 the study area is 221 and 187 respectively in the year 2011, which has remained almost

231 unchanged in recent years, while that of  $PM_{2.5}$  is increasing rapidly from 57 stations in the year  
232 2011 to 117 in the year 2013. This is because the national air quality standard for  $PM_{2.5}$  in Japan  
233 was set in the year 2009 and the monitoring network was initially developed by placing  
234 continuous monitors at existing stations.

235 GA, SA and HGS algorithms were applied to three different network sizes; 1) 57 stations,  
236 which is consistent with the number of stations of  $PM_{2.5}$  in the year 2011, 2) 117 stations,  
237 consistent with that in the year 2013, 3) 150 stations, which is approximately 3 times as much as  
238 the first case. It should be noted that the locations of the existing stations were not considered  
239 because the application of the observation-free method is focused. Each of the algorithms are  
240 applied to each pollutant combined with each network size respectively, repeating each of these  
241 27 settings 30 times to capture random effects. The performances are compared in terms of the  
242 mean and the standard deviation (SD) of the fitness values of the respective 30 trials. The  
243 stopping parameters of iterations for GA, SA and HGS are set so that the total effort of all the  
244 three algorithms is similar for a fair comparison.

245 Data analysis was carried out using R statistical software 3.1.2 (R Core Team, 2014) with the  
246 package GA (Scrucca, 2013) for the basic scheme of GA algorithm, the package gstat (Pebesma,  
247 2004) for kriging.

248

## 249 **3. Results and discussion**

### 250 **3.1 Performance of the algorithms**

251 The mean and standard deviation (SD) of the fitness values obtained from 30 random trials of  
252 each algorithm for the three network sizes and the three pollutants are presented in Figure 6.  
253 Regarding the comparison between the performance of GA and SA, SA gives smaller means in  
254 all the cases. However, SD of GA is consistently smaller than that of SA. In the case of the  
255 network size of  $n=117$  for  $PM_{2.5}$ , for instance, SD of GA is as small as 36 % of that of SA. These  
256 results indicate that SA is able to find a better solution but is not as stable as GA.

257 The mean of HGS is between 95 and 96% of the means of GA in all the cases and SD is 26 -  
258 47% of that of GA. When compared to the results of SA, HGS gives smaller means than those of  
259 SA and the SD is as small as 13 - 29% of that of SA. This indicates that HGS outperforms GA  
260 and SA with a better search capability and stability.

261 The best network placements out of the 30 trials for each network size and pollutant obtained  
262 by HGS are presented in Figure 7. The selected sites in the best network designs are generally  
263 placed homogeneously. Although the distributions of the simulated values for the three pollutants  
264 have unique characteristics, the difference in the optimized network between the pollutants is  
265 small.

### 266 **3.2 Spatial characteristics of the resulting best networks**



267 As shown in Figure 7, there seems to be few differences between the optimal networks for the  
268 different pollutants. For a quantitative comparison, the fitness value of each pollutant is  
269 calculated using the optimized network obtained by HGS for all the pollutants. For instance,  
270 fitness values of PM<sub>2.5</sub> are calculated using optimized network for NO<sub>2</sub> and O<sub>3</sub>, which in turn are  
271 compared to that obtained using PM<sub>2.5</sub>-optimized network. The fitness value is computed for each  
272 solution of 30 trials. The obtained results are presented in Figure 8.

273 The fitness values for each pollutant generally lie in the same range, no matter which pollutant  
274 is used in the optimization. Thus, the network optimized for one pollutant also result in good  
275 fitness for the other pollutants, and the relative goodness of a network compared to others does  
276 not change between the pollutants.

277 As shown in Figure 4, the variograms of all said three pollutants used for the optimization are  
278 generally similar, spherical model with nugget very close to zero. Van Groenigen (2000) showed  
279 that the optimized networks obtained for linear, exponential and spherical variograms with a  
280 relatively low nugget were similar. The results in this study agree with this previous study.

### 281 **3.3 Interpolation error**

282 The monitoring network can be evaluated by how well it represents the spatial distribution of  
283 the concentrations in the domain of interest. More specifically, the concentration field is  
284 generated by ordinary kriging using the CTM simulated values at the selected sites in the

285 optimized network. The variogram is constructed using simulated values at the selected sites.  
286 Fifteen equally-spaced points on the outer side of the study area are included in the same way as  
287 described in the Section 2.2. The resulting map is compared to the CTM simulated field.

288 RMSE and  $R^2$  between the simulated values and kriged values for each set of trials are  
289 computed. The obtained RMSE is given in Figure 9. RMSE is less than  $0.6 \text{ } (\mu\text{g m}^{-3})$  for all the  
290 three algorithms for  $\text{PM}_{2.5}$ , less than 1.7 (ppb) for  $\text{NO}_2$  and less than 1.8 (ppb) for  $\text{O}_3$ . These  
291 RMSE values are 6, 8 and 9% relative to the ranges of the simulated values of  $\text{PM}_{2.5}$ ,  $\text{NO}_2$  and  $\text{O}_3$   
292 respectively. The difference in RMSE between the three algorithms is minute for each network  
293 size and pollutant.  $R^2$  is generally higher than 0.62 for all the cases and the difference in  $R^2$   
294 between the algorithms is also marginal (not shown here).

295 In addition, the kriged fields generated by the optimization result of the three algorithms for the  
296 same size and pollutant are similar to each other and generally capture the feature of the  
297 corresponding simulated fields. As an example, the error maps between the simulated and kriged  
298 fields obtained from the result of HGS for the size of  $n=57$  and for the three pollutants are given  
299 in Figure 10, where positive values represent overestimates and negative values represent  
300 underestimates. The errors for  $\text{PM}_{2.5}$  are close to zero, between  $-3.6$  and  $1.6 \text{ } (\mu\text{g m}^{-3})$ , over the  
301 region of interest. On the other hand, underestimates for  $\text{NO}_2$  and overestimates for  $\text{O}_3$  are found  
302 in limited areas such as agglomeration of Osaka. These areas correspond to the areas of the

303 highest concentrations of NO<sub>2</sub> or the lowest concentrations of O<sub>3</sub>. These under/overestimates are  
304 due to the lack of the selected site in such limited areas of high/low concentrations. However,  
305 under/overestimates are not significant for most of the region except for the limited areas. These  
306 results indicate that the optimized networks obtained by the three algorithms represent the spatial  
307 distribution of the concentrations in the region of interest sufficiently.

### 308 **3.4 Evaluation of the optimized network for daily and hourly values**

309 The monitoring networks should map not only annual mean concentrations but also daily or  
310 hourly mean concentrations well. In order to evaluate the optimized network versus daily and  
311 hourly means, interpolation errors (RMSE) between the simulated and kriged fields for each day  
312 and hour of the whole year are computed in the same way as described in the section 3.3.  
313 Variograms are derived in an automated way using package automap (Hiemstra et al., 2009),  
314 because manually constructing variograms for every single day or hour of the entire year would  
315 be too time consuming.

316 The resulted RMSE are given in Table 1. The finer the temporal scale is, the larger RMSE  
317 becomes, which may be due to less homogeneous spatial distribution of pollutants at a finer  
318 temporal scale. However, RMSE of all the pollutants for all network sizes are relatively small  
319 compared to the ranges of the simulated daily or hourly means of the corresponding pollutants.  
320 Although not every daily or hourly result is checked manually, these results demonstrates that the

321 optimized network obtained using the annual mean is also optimal, at least to some extent, to  
322 produce maps of daily or hourly mean concentrations.

323

#### 324 **4. Conclusions**

325 In this paper, the performance and the applicability of HGS that is a hybrid of GA and SA is  
326 evaluated and compared to the single GA and SA using CTM simulated values to derive  
327 variograms for the computation of the mean kriging variance as a design criterion for PM<sub>2.5</sub>, NO<sub>2</sub>  
328 and O<sub>3</sub> in the Kinki region of Japan for three network sizes. HGS consistently outperforms the  
329 rest of the two algorithms for all the network sizes and pollutants. This demonstrates that HGS  
330 successfully combines the advantages of GA and SA. The optimized network for one pollutant is  
331 optimal for other pollutants and the relative goodness of a network compared to others does not  
332 change between the pollutants. This is because the variograms of the pollutants are similar. This  
333 implies that only one pollutant might be necessary to be considered when a network with multiple  
334 pollutants of similar variograms is optimized simultaneously. The concentration fields are  
335 generated by ordinary kriging using the simulated values at the selected sites in the optimized  
336 networks obtained by GA, SA and HGS respectively, and compared to the simulated fields to  
337 obtain interpolation errors. The difference between the algorithms is minute and the optimized  
338 networks obtained by the three algorithms represent the spatial distribution of the concentrations

339 in the region of interest sufficiently. The optimized network to well map the annual mean is  
340 optimal, to some extent, to produce maps of daily or hourly mean concentrations.

341 It is demonstrated that HGS is a valid and feasible method for the optimization of an air  
342 monitoring network. The combination of HGS and CTM simulation to derive a variogram for a  
343 criterion calculation is of benefit especially when a network with a limited number of stations or  
344 network under development is considered, and provide information of vital use on the air  
345 monitoring network optimization.

346

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349

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406



407 **Figure Captions**

408 Figure 1. The area of interest and the spatial distribution of PM<sub>2.5</sub>, NO<sub>2</sub> and O<sub>3</sub> concentrations  
409 obtained by CTM for the Japanese fiscal year 2010. The values for PM<sub>2.5</sub> and NO<sub>2</sub> are annual  
410 means and that for O<sub>3</sub> is annual mean of daily maximum 8-hr means. The circles represent the 15  
411 extra points used to derive variograms. Units are  $\mu\text{g m}^{-3}$  for PM<sub>2.5</sub> and ppb for NO<sub>2</sub> and O<sub>3</sub>.

412

413 Figure 2. The scatter plots of the simulated and observed values for annual means of PM<sub>2.5</sub> and  
414 NO<sub>2</sub>, and annual means of daily maximum 8-hr mean concentrations for O<sub>3</sub>. Units are  $\mu\text{g m}^{-3}$  for  
415 PM<sub>2.5</sub> and ppb for NO<sub>2</sub> and O<sub>3</sub>.

416

417 Figure 3. Histograms of the simulated values for PM<sub>2.5</sub>, NO<sub>2</sub> and O<sub>3</sub>. Units are  $\mu\text{g m}^{-3}$  for PM<sub>2.5</sub>  
418 and ppb for NO<sub>2</sub> and O<sub>3</sub>.

419

420 Figure 4. The variograms derived for PM<sub>2.5</sub>, NO<sub>2</sub> and O<sub>3</sub>.

421

422 Figure 5. The flowchart of the HGS algorithm.

423

424 Figure 6. The mean fitness values obtained from 30 random trials of each algorithm for PM<sub>2.5</sub> (in  
425 the left row panels), NO<sub>2</sub> (in the middle row panels) and O<sub>3</sub> (in the right row panels) for the  
426 network size of n=57 (in the upper column panels), n=117 (in the middle column panels) and  
427 n=150 (in the lower column panel). The fitness values are normalized by the mean values of GA  
428 of the corresponding network size and pollutant. The error bars represent SD.

429

430 Figure 7. The optimized network by HGS for PM<sub>2.5</sub> (in the left row panels), NO<sub>2</sub> (in the middle  
431 row panels) and O<sub>3</sub> (in the right row panels) for the network size of n=57 (in the upper column  
432 panels), n=117 (in the middle column panels) and n=150 (in the lower column panel). The  
433 crosses represent the selected sites. The circles represent the 15 extra points used to derive  
434 variograms.

435

436 Figure 8. The mean fitness values of a set of 30 trials for PM<sub>2.5</sub> (in the left row panels), NO<sub>2</sub> (in  
437 the middle row panels) and O<sub>3</sub> (in the right row panels) obtained using the network optimized for  
438 each pollutant for the network size of n=57 (in the upper column panels), n=117 (in the middle  
439 column panels) and n=150 (in the lower column panel). The top legend refers to the pollutant for  
440 which fitness value is computed using the optimized network of the three pollutants and the  
441 bottom legend refers to the pollutant for which the network is optimized for. The shaded areas

442 indicate that the fitness values are calculated for a pollutant using the optimized network of the  
443 same pollutant. The fitness values in each panel are normalized by mean values of PM<sub>2.5</sub>-  
444 optimized network in the same panel. The error bars represent SD.

445

446 Figure 9. RMSE between CTM simulated field and kriged field with the simulated values at the  
447 selected sites of the optimized network of each set of 30 trials by GA, SA and HGS for the  
448 network size of n=57, 117 and 150, and for PM<sub>2.5</sub>, NO<sub>2</sub> and O<sub>3</sub>. The error bars represent SD.

449 Units of RMSE for PM<sub>2.5</sub> are  $\mu\text{g m}^{-3}$  and ppb for NO<sub>2</sub> and O<sub>3</sub>.

450

451 Figure 10. The error map between the CTM simulated fields and the kriged fields with the  
452 simulated values at the selected sites of the optimized network of a) PM<sub>2.5</sub>, b) NO<sub>2</sub> and c) O<sub>3</sub> by  
453 HGS for the network size of n=57. The positive values represent overestimates and the negative  
454 values represent underestimates. The crosses represent the selected sites. The circles represent the  
455 15 extra points used to derive variograms. Units of concentrations are  $\mu\text{g m}^{-3}$  for PM<sub>2.5</sub> and ppb  
456 for NO<sub>2</sub> and O<sub>3</sub>.

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