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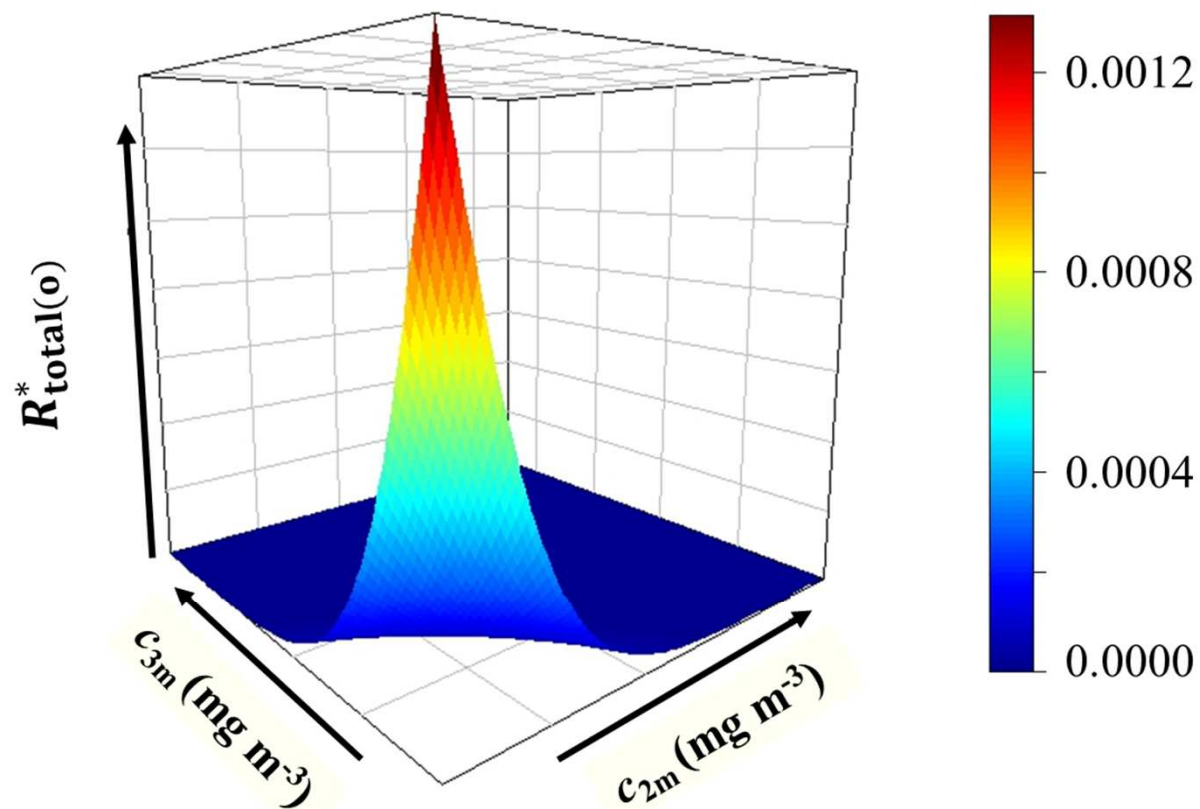
Dependence of the total risk of overestimation $R^*_{\text{total(o)}}$ of suspended particulate matter concentration in ambient air on the measurement results c_{im} in proximity to the three quarries (c_{1m} = 0.250 mg m^{-3}; c_{2m} and c_{3m} are varying from 0.210 to 0.300 mg m^{-3}).

Risk of a false decision on conformity of an environmental compartment due to measurement uncertainty of concentrations of two or more pollutants

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1. Introduction

Actual ('true') concentration c_i of the i-th pollutant, $i = 1, 2, \ldots, n,$ in an environmental compartment, e.g. ambient air (Duursma and Carroll (1996); TIMBRE project, Online), should not exceed a regulation or legal tolerance upper limit T_{Ui}. 'Concentration' is used here as a generic term (Cvitaš, 1996; Tolhurst, 2005; Fuentes-Arderiu, 2013). Comparing a chemical analytical test/measurement result c_{im} of the i-th pollutant concentration with the T_{Ui} value, one should decide whether the compartment conforms to the regulation or not. Since any result c_{im} has an associated measurement uncertainty (Ellison and Williams, 2012; Magnusson et al., 2012), several kinds of risk of a false decision on conformity of the compartment may arise.

The probability of a decision that the actual pollutant concentration does not exceed the limit since $c_{im} \le T_{Ui}$, when it is not correct (i.e. $c_i > T_{Ui}$), is named 'consumer's risk'. The 'consumer' in the present paper is a habitant whose quality of life (including health) depends on adequate control of the pollutant. Thus, the consumer's risk is the probability of underestimation of c_i due to measurement uncertainty associated with c_{im}.

On the other hand, the probability of falsely rejecting the decision on conformity of the compartment to the regulation (i.e. $c_{im} > T_{Ui}$ when $c_i \le T_{Ui}$) is the 'producer's risk'. The 'producer' here is a plant or another organization – a source of the environment pollution, obliged to pay a fine and/or to invest money for an unnecessary reduction of the pollutant concentration in the case of false nonconformity. The producer's risk is therefore the probability of overestimation of c_i due to measurement uncertainty in c_{im}.

For a specified compartment, e.g. ambient air in a certain location at a certain time, such risks are referred to as the 'specific consumer's risk' of underestimation $R^*_{ci(u)}$ and the 'specific

46   producer's risk' of overestimation $R^*_{ci(o)}$ for i-th particular pollutant concentration. The risks of

47   incorrect conformity assessment of a compartment randomly drawn from a statistical population

48   of such compartments are the 'global consumer's risk' of underestimation $R_{ci(u)}$ and the 'global

49   producer's risk' of overestimation $R_{ci(o)}$, respectively, as they characterize the environmental

50   quality globally. Evaluation of the particular risks (both specific and global) is described in the

51   JCGM 106 (2012) based on a Bayesian approach to conformity assessment.

52   However, when concentrations of two or more pollutants are controlled, pollutant-by-

53   pollutant evaluation of the risks is not complete in general, as it does not give an answer to the

54   question of the probability of a false decision on the overall compartment conformity. If

55   conformity assessment for each i-th pollutant concentration of a compartment is successful, i.e.

56   the particular specific R^*_{ci} or global R_{ci} risks of both under- and overestimation are small enough,

57   the total probability of a false decision concerning conformity of the compartment as a whole

58   (the *total* specific R^*_{total} or *total* global R_{total} risk) might still be significant.

59   A scheme summarizing the used terminology is shown in Fig. 1, where the particular risks

60   described in the JCGM 106 (2012) are shown at the top of the scheme. The *total risk evaluation*,

61   as the task of the IUPAC Project (2016), is highlighted by an ellipse at the bottom of the scheme.

62   Using the law of total probability for the case of independent quantities (pollutant

63   concentration values and corresponding measurement results) the total risk of underestimation

64   can be evaluated as a combination of the particular risks (Kuselman et al., 2017a). For example,

65   for three pollutions $i = 1, 2, 3$, assuming independent actual values of each pollutant

66   concentration c_i and independent corresponding measurement results c_{im}, the total specific risk

67   of underestimation is:

68

69 $$R^*_{\text{total(u)}} = R^*_{c1(u)} + R^*_{c2(u)} + R^*_{c3(u)} - R^*_{c1(u)}R^*_{c2(u)} - R^*_{c1(u)}R^*_{c3(u)} - R^*_{c2(u)}R^*_{c3(u)} +$$

70 $$R^*_{c1(u)}R^*_{c2(u)}R^*_{c3(u)}. \tag{1}$$

71

72 E.g., for all the particular specific risks $R^*_{ci(u)} = 0.05$, the total specific risk by formula (1) is

73 $R^*_{\text{total}} = 0.14$. Total global risk of underestimation for the three pollutants is:

74

75 $$R_{\text{total(u)}} =$$

76 $$P(C_2)P(C_3)R_{c1(u)} + P(C_1)P(C_3)R_{c2(u)} + P(C_1)P(C_2)R_{c3(u)} - P(C_3)R_{c1(u)}R_{c2(u)} -$$

77 $$P(C_2)R_{c1(u)}R_{c3(u)} - P(C_1)R_{c2(u)}R_{c3(u)} + R_{c1(u)}R_{c2(u)}R_{c3(u)}, \tag{2}$$

78

79 where $P(C_i)$ is the probability that a measurement result c_{im} is acceptable, i.e. $c_{im} \leq T_{Ui}$. For

80 example, for the particular risks $R_{ci} = 0.05$ and probabilities $P(C_i) = 0.90$ for all i, formula (2)

81 gives $R_{\text{total}} = 0.12$.

82   General expressions for evaluating the total risk of underestimation for any number n of the

83 material components (or pollutants of an environmental compartment) are also provided in the

84 mentioned above reference. Treatment of correlated measurement results for total risk evaluation is

85 discussed in the paper by Kuselman et al. (2017b).

86   In the present paper, the total risk of overestimation (producer's risk) is formulated in the same

87 Bayesian framework for uncorrelated test results as it was applied in the previous work (Kuselman

88 et al., 2017a) for underestimation (consumer's risk). Core code developed in R programming

89 environment (the R project, Online) for corresponding calculations is also provided. As a case

90 study, total risk values are calculated for conformity assessment of concentration of total suspended

91 particulate matter (TSPM) in ambient air from three independent stone quarries in Israel. In this

92   study TSPM contributed by the i-th quarry, $i = 1, 2, 3$, is considered as the i-th pollutant. While

93   particular risk values of false decisions on conformity of the i-th TSPM concentration, evaluated

94   earlier (Kuselman et al., 2012a), were related to each i-th pollutant (i-th quarry) separately, the total

95   risk values discussed below allow characterization of conformity of the TSPM concentration in the

96   region of the quarries as a whole. That is important as for the Regulator (the Ministry of

97   Environmental Protection, Online) protecting the inhabitants' quality of life in the area surrounding

98   the quarries, as for the Manufacturers Association (Online) acting in the interests of the stone

99   producers in the country.

100

101   **2. Methods**

102

103   **2.1. Raw data**

104

105   *2.1.1. Test method and likelihood functions*

106

107   A measured TSPM concentration in ambient air $c_{i\text{m}}$, mg m^{-3}, is an averaged mass of particles

108   with aerodynamic diameters of 100 μm or less collected from the air drawn through a filter in a

109   high-volume sampler over the sampling period of the test in proximity to the i-th stone quarry.

110   The testing was organized at a distance of (1-3) km from each quarry during the quarry' work.

111   Each test lasted 24 hours for collection of particles from about 2000 m^3 of air (EPA IO-2.1,

112   1999). The distribution of the test/measurement results $c_{i\text{m}}$ at the actual concentration c_i was

113   found to be normal with standard deviation equal to the standard measurement uncertainty $u_i =$

114   0.07 c_{im} and mean equal to c_i (Kuselman et al., 2012a). Corresponding likelihood functions are

115   normal probability density functions (pdfs):

116

$$f(c_{im}|c_i) = \frac{1}{u_i\sqrt{2\pi}}\exp\left[-\frac{(c_{im} - c_i)^2}{2u_i^2}\right]. \tag{3}$$

117

118   *2.1.2. Database and prior distributions of actual* concentration *values*

119

120   The database of 496 test results obtained during a year and described in the work of

121   Kuselman et al. (2012a) is considered again in the present paper. On the basis of the analysis of

122   variances (ANOVA), it was shown that the wind from the desert did not influence the test results

123   significantly, whereas anthropogenic contributions to TSPM concentration were dominant. No

124   correlation among test results for different quarries was observed. The theoretical distributions of

125   actual values of TSPM concentration c_i, fitting successfully the data collected close to quarry i,

126   were lognormal distributions, used in the following as prior pdfs:

127

$$f(c_i) = \frac{1}{c_i\sigma_i\sqrt{2\pi}}\exp\left[-\frac{(\ln c_i - \mu_i)^2}{2\sigma_i^2}\right], \tag{4}$$

128

129   where standard deviations σ_i and means μ_i are for the first quarry (i = 1) 0.434 and -2.326,

130   respectively, on the logarithmic scale; for the second quarry (i = 2) they are 0.280 and -2.031,

131   respectively; and for the third quarry σ_3 = 0.403 and μ_3 = -2.338.

132

133   **2.2. Regulation and acceptance limits**

134

135   There are national regulations of ambient air quality including upper regulation limits $T_{\mathrm{U}i}$ for

136   TSPM concentration depending on the period of sampling. In Israel, $T_{\mathrm{U}i} = 0.200$ mg m^{-3} for 24

137   hours, i.e. the same limit value is valid for any location in the country, also close to the i-th

138   quarry.

139   Besides the regulation limit, a lower/stricter acceptance limits A_i could be applied for the test

140   results with the purpose of decreasing the underestimation (inhabitant's) risks due to

141   measurement uncertainty u_i. In such a case, the decision rules (is the air conforming or not?) are

142   based on comparing the test results with the relevant i-th acceptance limit (JCGM 106, 2012;

143   Ellison and Williams, 2007). The acceptance limits in the present study are taken as coincidental

144   with the regulation limits.

145

146   **2.3. Particular risks of under- and overestimation**

147

148   *2.3.1. Particular specific risks*

149

150   The particular specific risks of the pollutant concentration under- and overestimation are

151   respectively

152

153    $$R^*_{ci(u)} = \int_{T_{\mathrm{U}i}}^{\infty} f(c_i|c_{im})\,\mathrm{d}c_i, \text{ for } c_{im} \leq T_{\mathrm{U}i}, \text{ and} \tag{5a}$$

154

155    $$R^*_{ci(o)} = \int_{0}^{T_{\mathrm{U}i}} f(c_i|c_{im})\,\mathrm{d}c_i, \text{ for } c_{im} > T_{\mathrm{U}i}, \tag{5b}$$

156

157   where $f(c_i|c_{im})$ is the posterior pdf for the actual value of the TSPM concentration c_i

158   contributed by the i-th quarry, given the measurement result near the quarry c_{im}. From Bayes

159   Law the posterior pdf is

160

161   $f(c_i|c_{im}) = f(c_{im}|c_i)f(c_i)/\int_{-\infty}^{\infty} f(c_{im}|c_i)f(c_i)\,dc_i,$ (5c)

162

163   where $f(c_{im}|c_i)$ is the likelihood function by eqn (3) and $f(c_i)$ is the prior pdf by eqn (4).

164

165   *2.3.2. Particular global risks*

166

167   The global risks of c_i under- and overestimation related to the TSPM regulation limit T_{Ui}, are

168   respectively

169

170   $R_{ci(u)} = \int_{T_{Ui}}^{\infty} \int_0^{T_{Ui}} f(c_{im}|c_i)f(c_i)\,dc_{im}dc_i,$ (6a)

171

172   $R_{ci(o)} = \int_0^{T_{Ui}} \int_{T_{Ui}}^{\infty} f(c_{im}|c_i)f(c_i)\,dc_{im}dc_i.$ (6b)

173

174   *2.3.3. Probabilities of an acceptable test result and a conforming actual concentration value*

175

176   Probability $P(C_i)$ of a conforming test/measurement result for the i-th pollutant ($c_{im} \leq A_i =$

177   T_{Ui}) is calculated by marginalization of the joint pdf of the measurement results and the actual

178   values of TSPM concentration:

179

$$P(\mathrm{C}_i) = \int_0^\infty \int_0^{T_{\mathrm{U}i}} f(c_{im}|c_i) f(c_i) \, \mathrm{d}c_{im} \mathrm{d}c_i \, . \tag{7a}$$

Probability $P(\overline{\mathrm{B}}_i)$ that the actual concentration value for the i-th pollutant is conforming $(c_i \leq T_{\mathrm{U}i})$ is calculated as:

$$P(\overline{\mathrm{B}}_i) = \int_0^{T_{\mathrm{U}i}} f(c_i) \, \mathrm{d}c_i \, . \tag{7b}$$

Note that the probability $P(\overline{\mathrm{B}}_i)$ of a conforming actual (true) value c_i in eqn (7b) does not depend on the measurement result c_{im} by definition. However, the vice versa holds: probability $P(\mathrm{C}_i)$ of a conforming measurement result c_{im} by eqn (7a) does depend on the relevant actual value c_i.

3. Modeling and calculation

3.1. Total risks of overestimation

3.1.1. Events

Define the following events possible during testing concentrations of two or more pollutants in an environmental compartment:

- $\overline{\mathrm{B}}_i$: the actual concentration c_i of pollutant i does not exceed its regulation limit $T_{\mathrm{U}i}$; probability of this event $P(\overline{\mathrm{B}}_i)$ is defined by formula (7b).

202   ▪ $\overline{B}$: the actual concentration values c_i for any i do not exceed their own regulation limits

203   T_{Ui}, $\overline{B} = \overline{B}_1 \cap \overline{B}_2 \cap ... \cap \overline{B}_n$; probability of this event is $P(\overline{B}) = \prod_{i=1}^{n} P(\overline{B}_i)$, if $\overline{B}_i$ are

204   mutually independent.

205   ▪ B_i: the actual concentration c_i of pollutant i exceeds T_{Ui}, i.e. violates it; probability of this

206   event is $P(B_i) = 1 - P(\overline{B}_i)$.

207   ▪ B: the actual concentration values c_i of one or more pollutants exceed their regulation

208   limits T_{Ui}, $B = B_1 \cup B_2 \cup ... \cup B_n$; probability of this event is $P(B) = 1 - P(\overline{B}) = 1 -$

209      $\prod_{i=1}^{n} P(\overline{B}_i)$.

210   ▪ C_i: the test result c_{im} for i-th pollutant does not exceed its acceptance limit A_i; probability

211   of this event $P(C_i)$ is defined by formula (7a).

212   ▪ C: the test results c_{im} for any i do not exceed their own acceptance limits A_i, $C = C_1 \cap$

213      $C_2 \cap ... \cap C_n$; probability of this event is $P(C) = \prod_{i=1}^{n} P(C_i)$, if C_i are mutually

214   independent.

215   ▪ $\overline{C}_i$: the test result c_{im} for i-th pollutant exceeds its acceptance limit A_i, i.e. such c_{im} is an

216   out-of-specification test result (Kuselman et al., 2012b) as $A_i = T_{Ui}$ in the present study;

217   probability of this event is $P(\overline{C}_i) = 1 - P(C_i)$.

218   ▪ $\overline{C}$: one or more test results c_{im} exceed their own A_i, $\overline{C} = \overline{C}_1 \cup \overline{C}_2 \cup ... \cup \overline{C}_n$; probability of

219   this event is $P(\overline{C}) = 1 - P(C) = 1 - \prod_{i=1}^{n} P(C_i)$.

220

221   *3.1.2. Total specific risk*

222

223   When a specified environmental compartment is tested concerning concentrations of three

224   pollutants, the total specific risk of overestimation $R_{\text{total}(o)}^{*}$ is the probability that the actual

225   concentrations of all pollutants in this compartment conform to their regulation limits ($\overline{\overline{B}} = \overline{B}_1 \cap$

226   $\overline{B}_2 \cap \overline{B}_3$), whereas one or more test/measurement results c_{1m}, c_{2m} and c_{3m} exceed their

227   acceptance limits. This event can occur when:

228   a) Just one measurement result out of the three, for example c_{1m} without losing generality,

229   exceeds its acceptance limit, while the actual concentration c_1 does not exceed the

230   regulation limit. In this case, the actual concentration c_1 will be overestimated. Hence, the

231   total risk that the compartment is falsely considered as not conforming is equal to the

232   particular specific risk concerning the first pollutant: $R^*_{total(o)} = P(\overline{B}_1|c_{1m})$.

233   b) Two measurement results, e.g. c_{1m} and c_{2m}, exceed their acceptance limits. The total risk

234   is $R^*_{total(o)} = P(\overline{B}_1 \cap \overline{B}_2|c_{1m}, c_{2m})$.

235   c) All the three measurement results exceed their acceptance limits. The total risk is

236      $R^*_{total(o)} = P(\overline{\overline{B}}|c_{1m}, c_{2m}, c_{3m}) = P(\overline{B}_1 \cap \overline{B}_2 \cap \overline{B}_3|c_{1m}, c_{2m}, c_{3m})$.

237   If the events $\overline{B}_i$ are conditionally independent, i.e. independent of the measurement results c_{im},

238   the total specific risk in each of the three considered situations is, respectively:

239

240   a) $R^*_{total(o)} = P(\overline{B}_1|c_{1m})$,   (8a)

241   b) $R^*_{total(o)} = \prod_{i=1}^{2} P(\overline{B}_i|c_{im})$,   (8b)

242   c) $R^*_{total(o)} = \prod_{i=1}^{3} P(\overline{B}_i|c_{im})$,   (8c)

243

244   where $P(\overline{B}_i|c_{im}) = R^*_{ci(o)}$ by formula (5b).

245   For any number n of pollutants, $v \leq n$ of which are characterized by the measurement results

246   exceeding their acceptance limits, the total specific risk of overestimation is

247

248 $$R^*_{\text{total(o)}} = \prod_{i=1}^{v} R^*_{ci(o)}. \tag{9}$$

249

250   Note again that $R^*_{ci(o)}$ in eqn (9) are related to the out-of-specification measurement results of

251   concentrations of the pollutants, sorted as the first v from all n pollutants under control.

252   From eqn (9) it follows that any one of v particular specific risk of overestimation $R^*_{ci(o)}$ equal

253   to zero will lead to $R^*_{\text{total(o)}} = 0$. That occurs when the actual concentration of the i-th pollutant

254   c_i exceeds/violates the regulation limit unquestionably ($c_i > T_{Ui}$) at a given measurement result

255    $c_{im} > T_{Ui}$ for this pollutant. In such a case, which does not depend on measurement results of

256   concentrations of the other pollutants, the compartment as a whole is certainly not conforming.

257   Therefore, the producer(s) should take action to reduce the i-th pollutant concentration and/or to

258   pay a fine.

259   In the opposite case of a particular specific risk value $R^*_{ci(o)} = 1$, although c_{im} exceeds its

260   acceptance limit, the actual concentration c_i certainly conforms. Such $R^*_{ci(o)}$ would not influence

261   the total specific risk $R^*_{\text{total(o)}}$ by eqn (9). In this case, the number n of pollutants is *de-facto*

262   decreased by one.

263   Another property of eqn (9) is reduction of $R^*_{\text{total(o)}}$ with increasing number v of pollutants

264   for which the measurement results are out-of-specification. The logic is that the more such

265   measurement results, the smaller is the total probability of the overestimation. Thus, the greater

266   is the probability that the compartment as a whole does not conform.

267   Note also that the model used in the work of Subaric-Leitis (2010) and adopted later in the

268   EURAMET guide (Pendrill et al., 2015) leads to an expression equivalent to eqn (9) when the

269   variables (concentrations of the pollutants in our task) are independent, hence validating the

270   model proposed in the present work.

271

272 *3.1.3. Total global risk*

273

274   Particular global risk $R_{ci(o)}$ of overestimation for the *i*-th pollutant ($i = 1, 2, 3$) is the

275 probability of false nonconformance when the corresponding test result exceeds its acceptance

276 limit A_i, while the actual value does not exceed the regulation limit T_{Ui}:

277

278     $R_{ci(o)} = P(\overline{B}_i \cap \overline{C}_i).$ (10)

279

280   The total global risk $R_{\text{total(o)}}$ of overestimation is the risk of having the actual concentrations

281 of the three pollutants within their regulation limits T_{Ui}, when at least one of test results are

282 outside its acceptance limits (that is outside the three-dimensional domain $A_1 \times A_2 \times A_3$), i.e.

283 $R_{\text{total(o)}} = P(\overline{B} \cap \overline{C})$, where

284

285 $\overline{B} \cap \overline{C} = \overline{B}_1 \cap \overline{B}_2 \cap \overline{B}_3 \cap (\overline{C}_1 \cup \overline{C}_2 \cup \overline{C}_3) = (\overline{B}_1 \cap \overline{B}_2 \cap \overline{B}_3 \cap \overline{C}_1) \cup (\overline{B}_1 \cap \overline{B}_2 \cap \overline{B}_3 \cap \overline{C}_2) \cup (\overline{B}_1 \cap$

286 $\overline{B}_2 \cap \overline{B}_3 \cap \overline{C}_3).$ (11)

287

288 The total global risk of overestimation is thus:

289

290 $R_{\text{total(o)}} = P(\overline{B}_1 \cap \overline{B}_2 \cap \overline{B}_3 \cap \overline{C}_1) + P(\overline{B}_1 \cap \overline{B}_2 \cap \overline{B}_3 \cap \overline{C}_2) + P(\overline{B}_1 \cap \overline{B}_2 \cap \overline{B}_3 \cap \overline{C}_3) - P(\overline{B}_1 \cap$

291 $\overline{B}_2 \cap \overline{B}_3 \cap \overline{C}_1 \cap \overline{C}_2) - P(\overline{B}_1 \cap \overline{B}_2 \cap \overline{B}_3 \cap \overline{C}_1 \cap \overline{C}_3) - P(\overline{B}_1 \cap \overline{B}_2 \cap \overline{B}_3 \cap \overline{C}_2 \cap \overline{C}_3) + P(\overline{B}_1 \cap$

292 $\overline{B}_2 \cap \overline{B}_3 \cap \overline{C}_1 \cap \overline{C}_2 \cap \overline{C}_3).$ (12)

293

294 Whenever $\overline{B}_1$, $\overline{B}_2$ and $\overline{B}_3$, as well as $\overline{C}_1$, $\overline{C}_2$, and $\overline{C}_3$, are mutually independent, events $\overline{B}_1 \cap \overline{C}_1$,

295 $\overline{B}_2 \cap \overline{C}_2$ and $\overline{B}_3 \cap \overline{C}_3$ are also independent and equation (12) can be rewritten using notation (10)

296 in the following way:

297

298 $R_{\text{total(o)}} =$

299 $P(\overline{B}_2)P(\overline{B}_3)R_{c1(o)} + P(\overline{B}_1)P(\overline{B}_3)R_{c2(o)} + P(\overline{B}_1)P(\overline{B}_2)R_{c3(o)} - P(\overline{B}_3)R_{c1(o)}R_{c2(o)} -$

300 $P(\overline{B}_2)R_{c1(o)}R_{c3(o)} - P(\overline{B}_1)R_{c2(o)}R_{c3(o)} + R_{c1(o)}R_{c2(o)}R_{c3(o)}.$ (13)

301

302 Note that eqn (13) is similar to eqn (2) for the total global risk of underestimation. However,

303 it involves probabilities of different events and different particular risks.

304 In general, for any number n of pollutants

305

306 $R_{\text{total(o)}} =$

307 $\sum_{i=1}^{n} \left(\prod_{l \neq i} P(\overline{B}_l) \right) R_{ci(o)} -$

308 $\sum_{i=1}^{n} \sum_{j>i} \left(\prod_{l \neq i,j} P(\overline{B}_l) \right) \left(\prod_{q=i,j} R_{cq(o)} \right) +$

309 $\sum_{i=1}^{n} \sum_{j>i} \sum_{k>j} \left(\prod_{l \neq i,j,k} P(\overline{B}_l) \right) \left(\prod_{q=i,j,k} R_{cq(o)} \right) + \cdots + (-1)^{n-2} \sum_{i=1}^{n} P(\overline{B}_i) \left(\prod_{q \neq i} R_{cq(o)} \right) +$

310 $(-1)^{n-1} \prod_{q=1}^{n} R_{cq(o)},$ (14)

311

312 where i, j, k, l and q are subscripts of the pollutant in the range $(1, \ldots, n)$.

313

314 **3.2. Calculation**

315

316　　　When the likelihood function is a normal distribution and the prior pdf is lognormal, the

317　　posterior pdf cannot be easily described by an analytical closed form. Therefore, the posterior

318　　pdf was obtained by numerical integration (and subsequent normalization) of the product of the

319　　prior and the likelihood. The under- and overestimation particular risks were calculated as the

320　　fraction of the (approximated) posterior pdf lying outside/inside the tolerance limit, respectively.

321　　　Core code developed in R programming environment for calculation of the risks is reported in

322　　Annex A. Calculation of total specific risks of under- and overestimation by eqns (1) and (8),

323　　respectively, using corresponding particular specific risk values by eqns (5), is shown in Section

324　　A-1. Time spent for calculation of the total specific risks with a regular PC (Intel® Core^{TM} i5-

325　　3470 Processor, CPU @ 3.20 GHz, Windows 7 Professional 64 bit) is about one second. While

326　　increasing (doubling, for example) the number of the involved components does not affect the

327　　calculation time, decreasing the numerical integration parameter (stepsize) from 0.001 to 0.0001,

328　　increases the execution time up to 6 seconds.

329　　　Calculation of total global risks of under- and overestimation by eqns (2) and (13),

330　　respectively, using particular global risk values by eqns (6), probabilities of conforming

331　　measurement results by eqn (7a) and probabilities of conforming actual concentration values by

332　　eqn (7b), is detailed in Section A-2. Time spent for calculation of the total global risks with the

333　　same PC is about 5 seconds. In this case, doubling the number of components doubles the

334　　required time, whereas decreasing the integration parameter (step) from 0.00001 to 0.000001

335　　increases the computational time up to about 37 seconds.

336

337　**4. Results and discussion**

338

4.1. Total specific risks of under- and overestimation

Dependence of the total specific risks of underestimation of TSPM concentration in air on the measurement results c_{im} is demonstrated in Fig. 2. A case when only the first quarry is active and the total risk $R^*_{\text{total(u)}}$ equals to the particular risk $R^*_{c1(u)}$, is shown in Fig. 2a by solid line 1. Dotted lines 3 and 2 point a measured TSPM concentration $c_{1m} = 0.194$ mg m^{-3} and corresponding risk value $R^*_{c1(u)} = 0.211$, as an instance. One can see in Fig. 2a that $R^*_{c1(u)}$ is close to zero (negligible) at $c_{1m} < 0.170$ mg m^{-3}, however significantly increasing with c_{1m} approaching the tolerance limit $T_{U1} = 0.200$ mg m^{-3}.

A case when only the second and the third quarries are active, is represented in Fig. 2b, where the total risk, $R^*_{\text{total(u)}}$, shown as a surface, depends on both c_{2m} and c_{3m} in the range [0.010, 0.200] mg m^{-3}. The surface lies mostly on the bottom of the three-dimensional region where $R^*_{\text{total(u)}}$ is close to zero, as in Fig. 2a, increasing with c_{2m} and c_{3m} approaching their tolerance limits $T_{U1} = T_{U2} = 0.200$ mg m^{-3}. When both c_{2m} and c_{3m} simultaneously approach 0.200 mg m^{-3}, this leads to a 'protuberance' in the total risk surface.

The same dependence of $R^*_{\text{total(u)}}$ on c_{2m} and c_{3m} is observed when all the three quarries are active simultaneously, but $c_{1m} < 0.170$ mg m^{-3}: the contribution of the particular risk $R^*_{c1(u)}$ to the total one in such a case is negligible as shown in Fig. 2a. For comparison, Fig. 2c illustrates a scenario when all the three quarries are active and $R^*_{\text{total(u)}}$ - the surface - is depending on c_{2m} and c_{3m} in the range [0.010, 0.200] mg m^{-3} as in Fig. 2b, whereas $c_{1m} = 0.194$ mg m^{-3}. Fig. 2c seems very similar to Fig. 2b. However, the color scales of the $R^*_{\text{total(u)}}$ surfaces are different, since the scale in Fig. 2c is greater because of the significant contribution of $R^*_{c1(u)} = 0.211$ at $c_{1m} = 0.194$ mg m^{-3} (indicated in Fig 2a by dotted lines).

362   Dependence of the total specific risks of overestimation of the actual TSPM concentration in

air on measurement results, when they are out-of-specification ($c_{im} > T_{Ui}$), is detailed in Fig. 3.

364   A case when only the first quarry is active, and the total risk $R^*_{total(o)}$ is equal to the particular

365   risk $R^*_{c1(o)}$, is shown in Fig. 3a by solid line 1. Dotted lines 3 and 2 point a measured TSPM

366   concentration $c_{1m} = 0.250$ mg m^{-3} and corresponding risk value $R^*_{c1(o)} = 0.008$, as an example.

367   Naturally, the risk of overestimation increases as c_{1m} approaches 0.200 mg m^{-3} (the tolerance

368   limit), and is close to zero for $c_{1m} > 0.260$ mg m^{-3}.

369   The case when only the second and the third quarries are active, as in Fig 2b, and $R^*_{total(o)}$

370   value depending on both c_{2m} and c_{3m} in the range $[0.210, 0.300]$ mg m^{-3}, is shown in Fig. 3b. The

371   maximum $R^*_{total(o)}$ value is observed as c_{2m} and c_{3m} near the tolerance limit simultaneously.

372   Fig. 3c illustrates a case when all the three quarries are active, as in Fig. 2c, but $c_{1m} = 0.250$

373   mg m^{-3}. The scale of the $R^*_{total(u)}$ surface, shown by the color bar, is two orders less than in Fig.

374   3b. The reason is that the total risk of overestimation, defined as a product of the three particular

375   risks, is influenced by the contribution of $R^*_{c1(o)} = 0.008$ at $c_{1m} = 0.250$ mg m^{-3} (indicated in Fig

376   3a by dotted lines). In other words, if an out-of-specification measurement result is significantly

377   greater than the tolerance limit, the probability of violation of the regulation is high and the

378   particular risk of overestimation is low. Therefore the total specific risk of overestimation is low

379   also.

380

381   **4.2. Total global risks of under- and overestimation**

382

383   The particular global risks of underestimation $R_{c1(u)} = 0.006$, $R_{c2(u)} = 0.010$ and $R_{c3(u)} =$

384   0.005 obtained here are equal to the values published earlier (Kuselman et al., 2012a). They are

385 used as a part of the validation process of the current calculations. The probabilities of

386 conforming measurement results are $P(C_1) = 0.949$, $P(C_2) = 0.929$ and $P(C_3) = 0.963$. The

387 total risk of underestimation, evaluated in the present work for the first time, is $R_{\text{total(u)}} = 0.019$,

388 hence greater than the particular risk contributed by each quarry.

389   The particular global risks of overestimation are $R_{c1(o)} = 0.007$, $R_{c2(o)} = 0.015$ and $R_{c3(o)} =$

390 0.006. They are also equal to those published by Kuselman et al. (2012a). The probabilities of

391 conforming actual concentration values calculated are $P(\overline{B}_1) = 0.951$, $P(\overline{B}_2) = 0.934$ and

392 $P(\overline{B}_3) = 0.965$. The total risk of overestimation, evaluated in the present work for the first time

393 as well, is $R_{\text{total(o)}} = 0.026$, again greater than each $R_{ci(o)}$.

394   The total risk of overestimation $R_{\text{total(o)}}$ exceeds the total risk of underestimation $R_{\text{total(u)}}$,

395 which implies that there is a reasonable balance between the requirements of an inhabitant's

396 quality of life and the producer's expenditure on environmental protection.

397

5. Conclusions

399

400   Quantification of risks of false decisions in conformity assessment of an environmental

401 compartment due to measurement uncertainty of concentrations of two or more pollutants, is

402 developed. Even if the assessment of conformity for each pollutant in the compartment is

403 successful, the total probability of a false decision concerning the compartment as a whole might

404 still be significant.

405   A model of the total probability of a false decision, formulated on the basis of the law of total

406 probability, is used for a study of test results of total suspended particulate matter concentration

407 in ambient air from three independent stone quarries in Israel. Total probabilities of

408 underestimation of the particulate matter concentration (total risk of the inhabitants) and

409 overestimation (total risk of the stone producers) are evaluated as a combination of the particular

410 risks of air conformity assessment near to each quarry.

411   It is shown that the total global risk of underestimation of the particulate matter concentration

412 is smaller than the total risk of its overestimation. That is a reasonable balance between the

413 requirements of an inhabitant's quality of life and the producer's expenditure on environmental

414 protection.

415

416 **Acknowledgment**

417

420

421 **Appendix A. Core of the R code**

422

423   **A-1. Calculation of the total specific risks**

424

```
425  ###############
426  # Specific risks #
427  ###############
428
429  # Input data for the quarries
430  mu1 = -2.326         # Prior location parameter for Q1
431  mu2 = -2.031         # Prior location parameter for Q2
432  mu3 = -2.338         # Prior location parameter for Q3
433  sigma1 = 0.434       # Prior scale parameter for Q1
434  sigma2 = 0.280       # Prior scale parameter for Q2
435  sigma3 = 0.403       # Prior scale parameter for Q3
```

```
436    Rsigmam = 0.07          # Relative measurement uncertainty
437    TU = 0.2                # Tolerance limit
438
439    # Settings for numerical integrations
440    stepsize <- 0.001
441    obsvalues = seq(0.01,TU,stepsize)
442    postmean = rep(0,length(obsvalues))
443    poststd  = rep(0,length(obsvalues))
444    Rspec1  = rep(0,length(obsvalues))
445    Rspec2  = rep(0,length(obsvalues))
446    Rspec3  = rep(0,length(obsvalues))
447    c = seq(0,0.5,stepsize)
448
449    ######################################################
450    # Consumer specific risk for each observed value in [0.01, TU]
451    # Normal Likelihood and Lognormal prior
452
453    # Q1
454    i = 1
455    prior <- dlnorm(c, meanlog = mu1, sdlog = sigma1)
456    logprior <- log(prior)
457    for(obs in obsvalues)
458    {
459     loglik <- dnorm(obs, mean = c, sd = Rsigmam*obs, log = T)
460     logpos <- logprior + loglik
461     posterior <- exp(logpos)
462     posterior <- posterior/(sum(posterior)*stepsize)
463     postmean[i] <- sum(posterior*c)*stepsize
464     postvar <- sum(posterior*(c^2))*stepsize-postmean[i]^2
465     poststd[i] = sqrt(postvar)
466     Rspec1[i] = stepsize*sum(posterior[c>TU])
467     i = i+1
468    }
469
470    # Q2
471    i = 1
472    prior <- dlnorm(c, meanlog = mu2, sdlog = sigma2)
473    logprior <- log(prior)
474    for(obs in obsvalues)
475    {
476     loglik <- dnorm(obs, mean = c, sd = Rsigmam*obs, log = T)
477     logpos <- logprior + loglik
478     posterior <- exp(logpos)
479     posterior <- posterior/(sum(posterior)*stepsize)
480     postmean[i] <- sum(posterior*c)*stepsize
481     postvar <- sum(posterior*(c^2))*stepsize-postmean[i]^2
```

```
482    poststd[i] = sqrt(postvar)
483    Rspec2[i] = stepsize*sum(posterior[c>TU])
484    i = i+1
485    }
486
487    # Q3
488    i = 1
489    prior <- dlnorm(c, meanlog = mu3, sdlog = sigma3)
490    logprior <- log(prior)
491    for(obs in obsvalues)
492    {
493    loglik <- dnorm(obs, mean = c, sd = Rsigmam*obs, log = T)
494    logpos <- logprior + loglik
495    posterior <- exp(logpos)
496    posterior <- posterior/(sum(posterior)*stepsize)
497    postmean[i] <- sum(posterior*c)*stepsize
498    postvar <- sum(posterior*(c^2))*stepsize-postmean[i]^2
499    poststd[i] = sqrt(postvar)
500    Rspec3[i] = stepsize*sum(posterior[c>TU])
501    i = i+1
502    }
503
504    # Total specific consumer risk for the particular case obs1=obs2=obs3
505    Rtotu = Rspec1 + Rspec2 + Rspec3 - Rspec1*Rspec2 - Rspec1*Rspec3 - Rspec2*Rspec3 +
506    Rspec1*Rspec2*Rspec3
507
508    ######################################################
509    # Producer specific risk for each observed value in [0.21, 0.3]
510
511    # Settings for numerical integrations
512    obsvalues = seq(0.21,0.3,stepsize)
513    postmean = rep(0,length(obsvalues))
514    poststd  = rep(0,length(obsvalues))
515    Rspec1  = rep(0,length(obsvalues))
516    Rspec2  = rep(0,length(obsvalues))
517    Rspec3  = rep(0,length(obsvalues))
518
519    # Q1
520    i = 1
521    prior <- dlnorm(c, meanlog = mu1, sdlog = sigma1)
522    logprior <- log(prior)
523    for(obs in obsvalues)
524    {
525    loglik <- dnorm(obs, mean = c, sd = Rsigmam*obs, log = T)
526    logpos <- logprior + loglik
527    posterior <- exp(logpos)
```

```
528    posterior <- posterior/(sum(posterior)*stepsize)
529    postmean[i] <- sum(posterior*c)*stepsize
530    postvar <- sum(posterior*(c^2))*stepsize-postmean[i]^2
531    poststd[i] = sqrt(postvar)
532    Rspec1[i] = stepsize*sum(posterior[c<=TU])
533    i = i+1
534    }
535
536    # Q2
537    i = 1
538    prior <- dlnorm(c, meanlog = mu2, sdlog = sigma2)
539    logprior <- log(prior)
540    for(obs in obsvalues)
541    {
542    loglik <- dnorm(obs, mean = c, sd = Rsigmam*obs, log = T)
543    logpos <- logprior + loglik
544    posterior <- exp(logpos)
545    posterior <- posterior/(sum(posterior)*stepsize)
546    postmean[i] <- sum(posterior*c)*stepsize
547    postvar <- sum(posterior*(c^2))*stepsize-postmean[i]^2
548    poststd[i] = sqrt(postvar)
549    Rspec2[i] = stepsize*sum(posterior[c<=TU])
550    i = i+1
551    }
552
553    # Q3
554    i = 1
555    prior <- dlnorm(c, meanlog = mu3, sdlog = sigma3)
556    logprior <- log(prior)
557    for(obs in obsvalues)
558    {
559    loglik <- dnorm(obs, mean = c, sd = Rsigmam*obs, log = T)
560    logpos <- logprior + loglik
561    posterior <- exp(logpos)
562    posterior <- posterior/(sum(posterior)*stepsize)
563    postmean[i] <- sum(posterior*c)*stepsize
564    postvar <- sum(posterior*(c^2))*stepsize-postmean[i]^2
565    poststd[i] = sqrt(postvar)
566    Rspec3[i] = stepsize*sum(posterior[c<=TU])
567    i = i+1
568    }
569
570    # Total specific producer risk for the particular case obs1=obs2=obs3
571    Rtoto = Rspec1*Rspec2*Rspec3
572
```

573   **A-2. Calculation of the total global risks**

574

575   #############
576   # Global risks #
577   #############
578
579   # Input data for the quarries
580   mu1 = -2.326   # Prior location parameter for Q1
581   mu2 = -2.031   # Prior location parameter for Q2
582   mu3 = -2.338   # Prior location parameter for Q3
583   sigma1 = 0.434   # Prior scale parameter for Q1
584   sigma2 = 0.280   # Prior scale parameter for Q2
585   sigma3 = 0.403   # Prior scale parameter for Q3
586   um = 0.07   # Relative measurement uncertainty
587   T = 0.2   # Tolerance limit
588   A = T   # Acceptance limit
589
590   # Consumer's risk Rc and the producer's risk Rp
591   # Normal Likelihood and Lognormal prior
592   # Initializations
593   step = 0.00001
594   etac = seq(T,10,step)   # Integral domain [T, infinity]
595   etap = seq(step,T,step)   # Integral domain [0, T]
596   etacinf = seq(step,10,step)   # Integral domain [0, infinity]
597
598   # Q1
599   ymeanlogQ1 = mu1
600   ystdlogQ1 = sigma1
601   RcQ1 = sum((pnorm((A-etac)/(um*etac)) - pnorm(-etac/(um*etac))) *
602   dlnorm(etac,ymeanlogQ1,ystdlogQ1) * step)
603   PC1 = sum((pnorm((A-etacinf)/(um*etacinf)) - pnorm(-etacinf/(um*etacinf))) *
604   dlnorm(etacinf,ymeanlogQ1,ystdlogQ1) * step)
605   RpQ1 = sum((1-pnorm((A-etap)/(um*etap))) * dlnorm(etap,ymeanlogQ1,ystdlogQ1) * step)
606   PBcompl1 = plnorm(T,ymeanlogQ1,ystdlogQ1)
607   c(RcQ1,RpQ1,PC1,PBcompl1)
608   # [1] 0.005769988 0.007368876 0.949038432 0.950637320
609
610   # Q2
611   ymeanlogQ2 = mu2
612   ystdlogQ2 = sigma2
613   RcQ2 = sum((pnorm((A-etac)/(um*etac)) - pnorm(-etac/(um*etac))) *
614   dlnorm(etac,ymeanlogQ2,ystdlogQ2) * step)
615   PC2 = sum((pnorm((A-etacinf)/(um*etacinf)) - pnorm(-etacinf/(um*etacinf))) *
616   dlnorm(etacinf,ymeanlogQ2,ystdlogQ2) * step)

```
617    RpQ2 = sum( (1- pnorm((A-etap)/(um*etap))) * dlnorm(etap,ymeanlogQ2,ystdlogQ2) * step)
618    PBcompl2 = plnorm(T,ymeanlogQ2,ystdlogQ2)
619    c(RcQ2,RpQ2,PC2,PBcompl2)
620    # [1] 0.01045913 0.01525355 0.92911792 0.93391234
621
622    # Q3
623    ymeanlogQ3 = mu3
624    ystdlogQ3 = sigma3
625    RcQ3 = sum( (pnorm((A-etac)/(um*etac)) - pnorm(-etac/(um*etac))) *
626    dlnorm(etac,ymeanlogQ3,ystdlogQ3) * step)
627    PC3 = sum( (pnorm((A-etacinf)/(um*etacinf)) - pnorm(-etacinf/(um*etacinf))) *
628    dlnorm(etacinf,ymeanlogQ3,ystdlogQ3) * step)
629    RpQ3 = sum( (1- pnorm((A-etap)/(um*etap))) * dlnorm(etap,ymeanlogQ3,ystdlogQ3) * step)
630    PBcompl3 = plnorm(T,ymeanlogQ3,ystdlogQ3)
631    c(RcQ3,RpQ3,PC3,PBcompl3)
632    # [1] 0.004602961 0.006233814 0.963053939 0.964684793
633
634    # TOTAL global consumer risk (underestimation risk)
635    c(PC1,PC2,PC3)
636    # [1] 0.9490384 0.9291179 0.9630539
637    c(RcQ1,RcQ2,RcQ3)
638    # [1] 0.005769988 0.010459133 0.004602961
639    Rtotu = PC2*PC3*RcQ1 + PC1*PC3*RcQ2 + PC1*PC2*RcQ3 - PC3*RcQ1*RcQ2 -
640    PC2*RcQ1*RcQ3 - PC1*RcQ2*RcQ3 + RcQ1*RcQ2*RcQ3
641    Rtotu #  0.01865286, for step = 0.00001
642
643    # TOTAL global producer risk (overestimation risk)
644    c(PBcompl1,PBcompl2,PBcompl3)
645    # [1] 0.9506373 0.9339123 0.9646848
646    c(RpQ1,RpQ2,RpQ3)
647    # [1] 0.007368876 0.015253553 0.006233814
648    Rtoto =  PBcompl2*PBcompl3*RpQ1 + PBcompl1*PBcompl3*RpQ2 +
649    PBcompl1*PBcompl2*RpQ3 - PBcompl3*RpQ1*RpQ2 - PBcompl2*RpQ1*RpQ3 -
650    PBcompl1*RpQ2*RpQ3 + RpQ1*RpQ2*RcQ3
651    Rtoto # 0.0259206, for step = 0.00001
652
```

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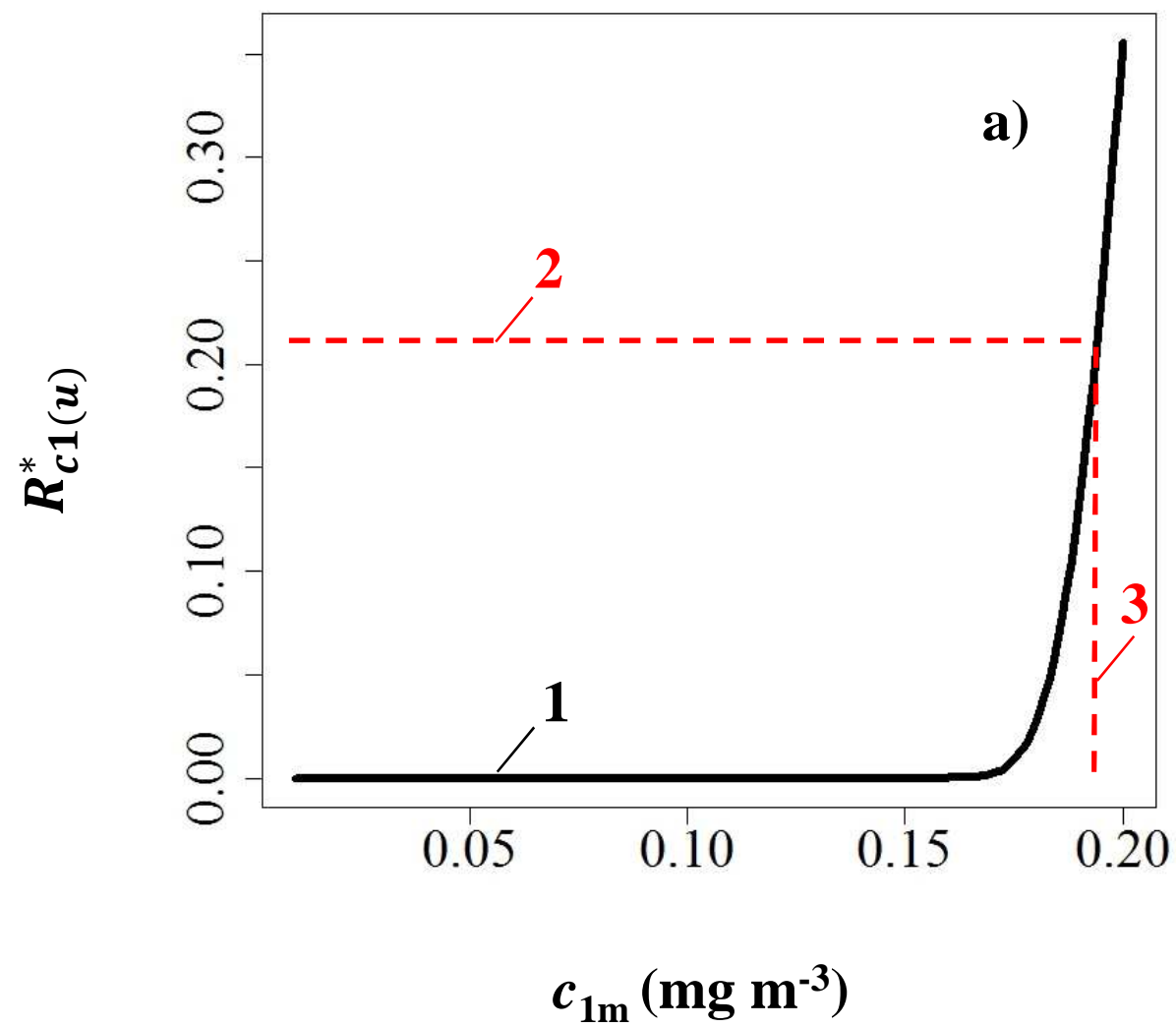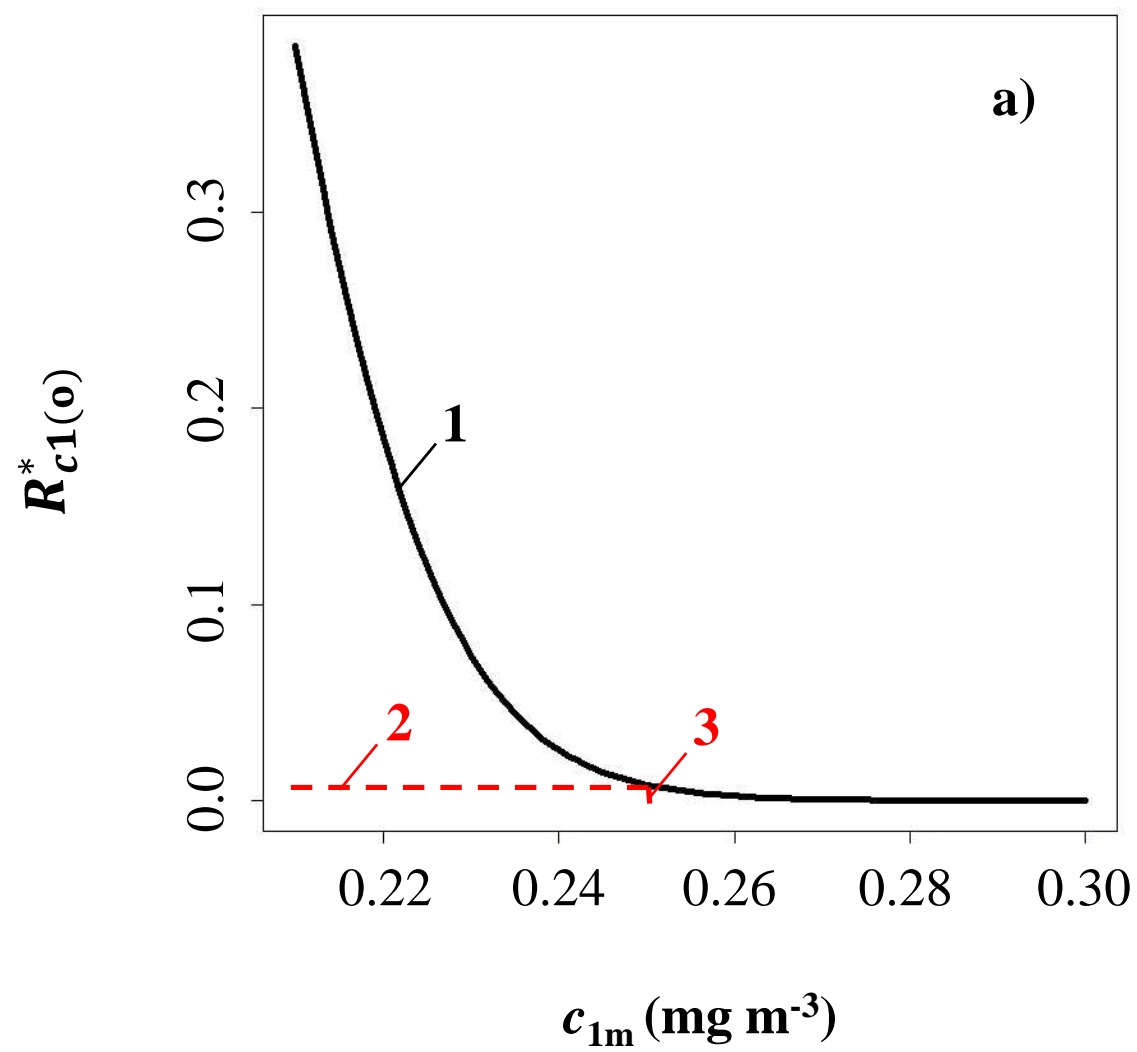708   sediments. Estuarine, Coastal and Shelf Science. 63, 665-673.

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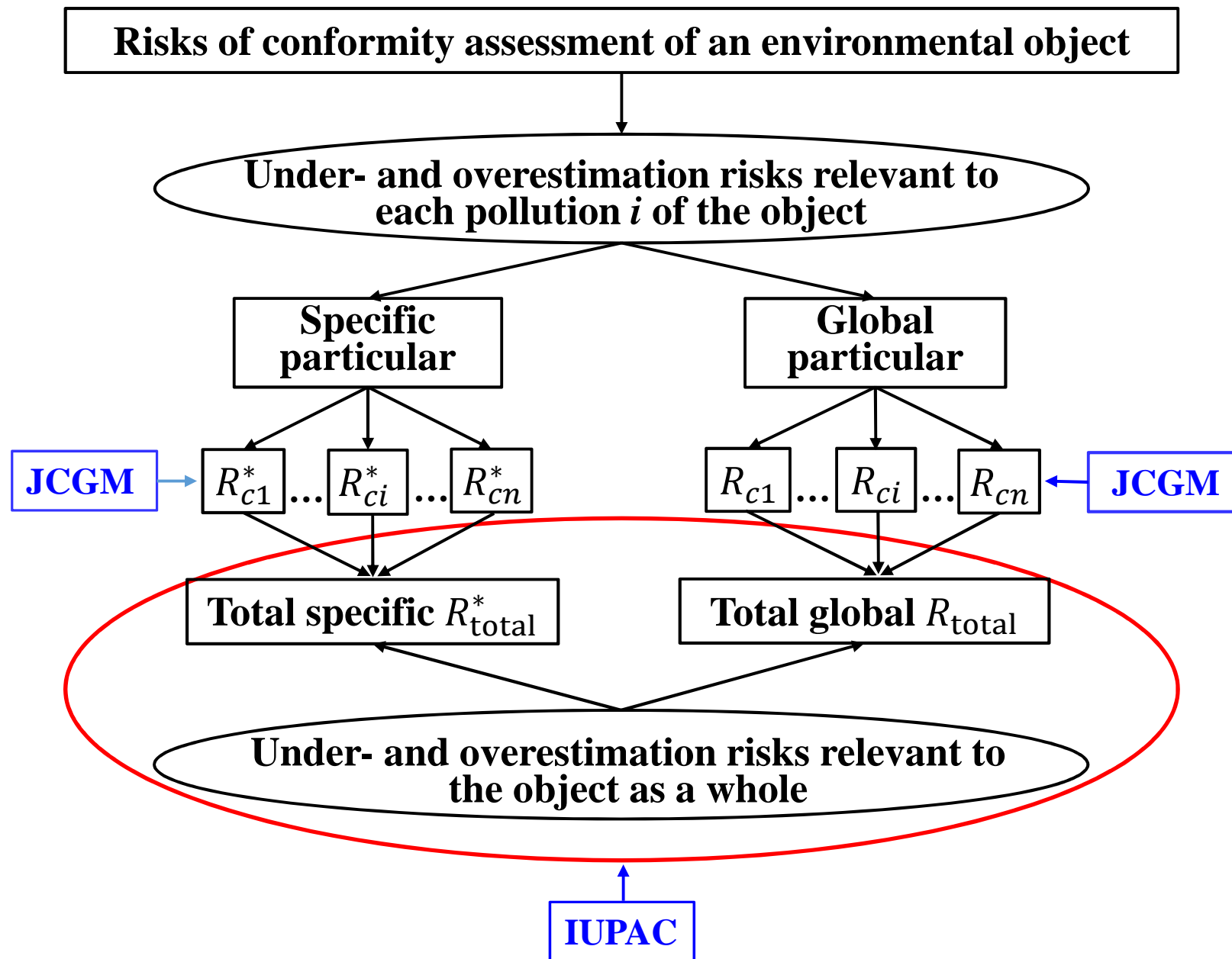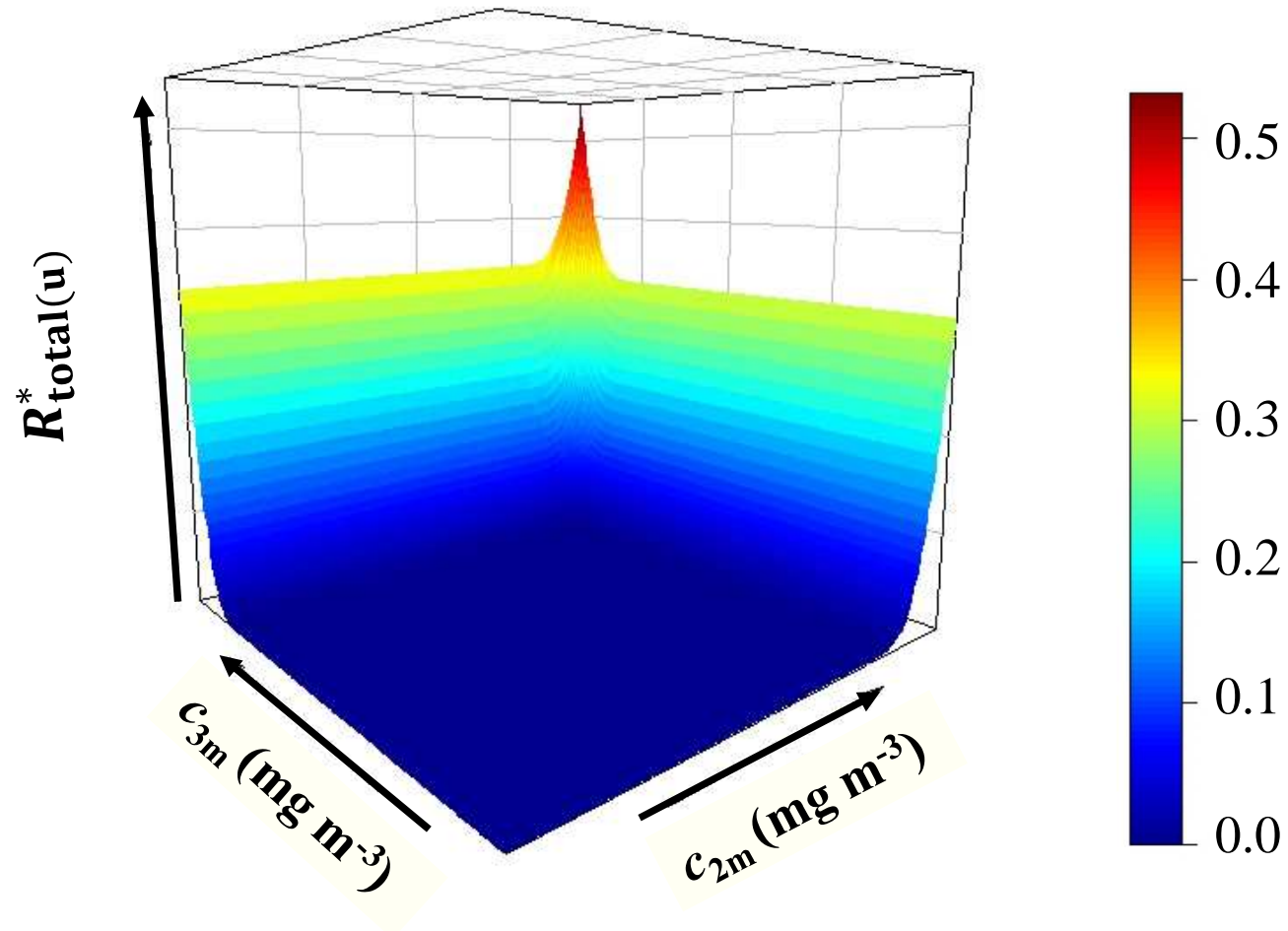
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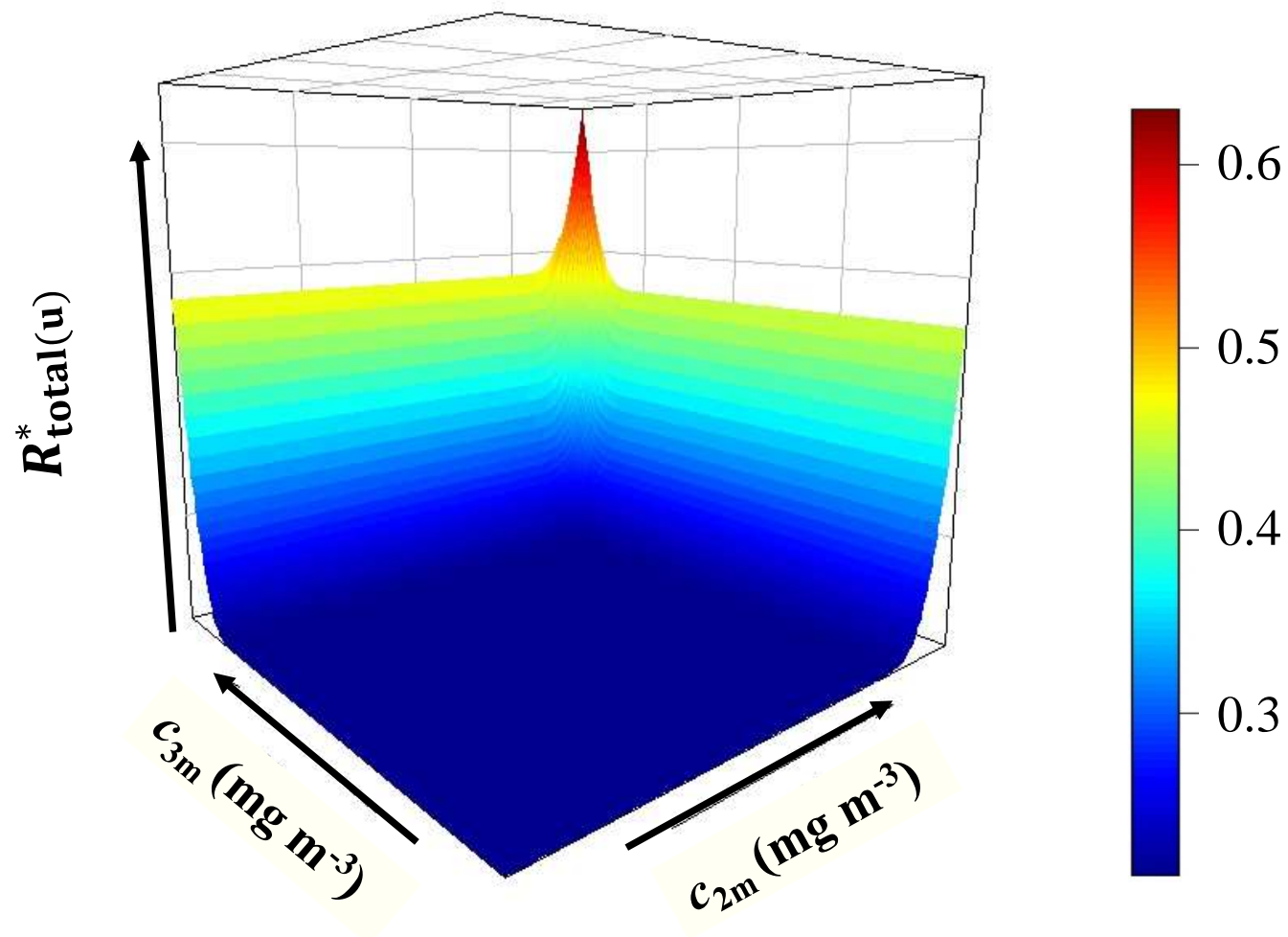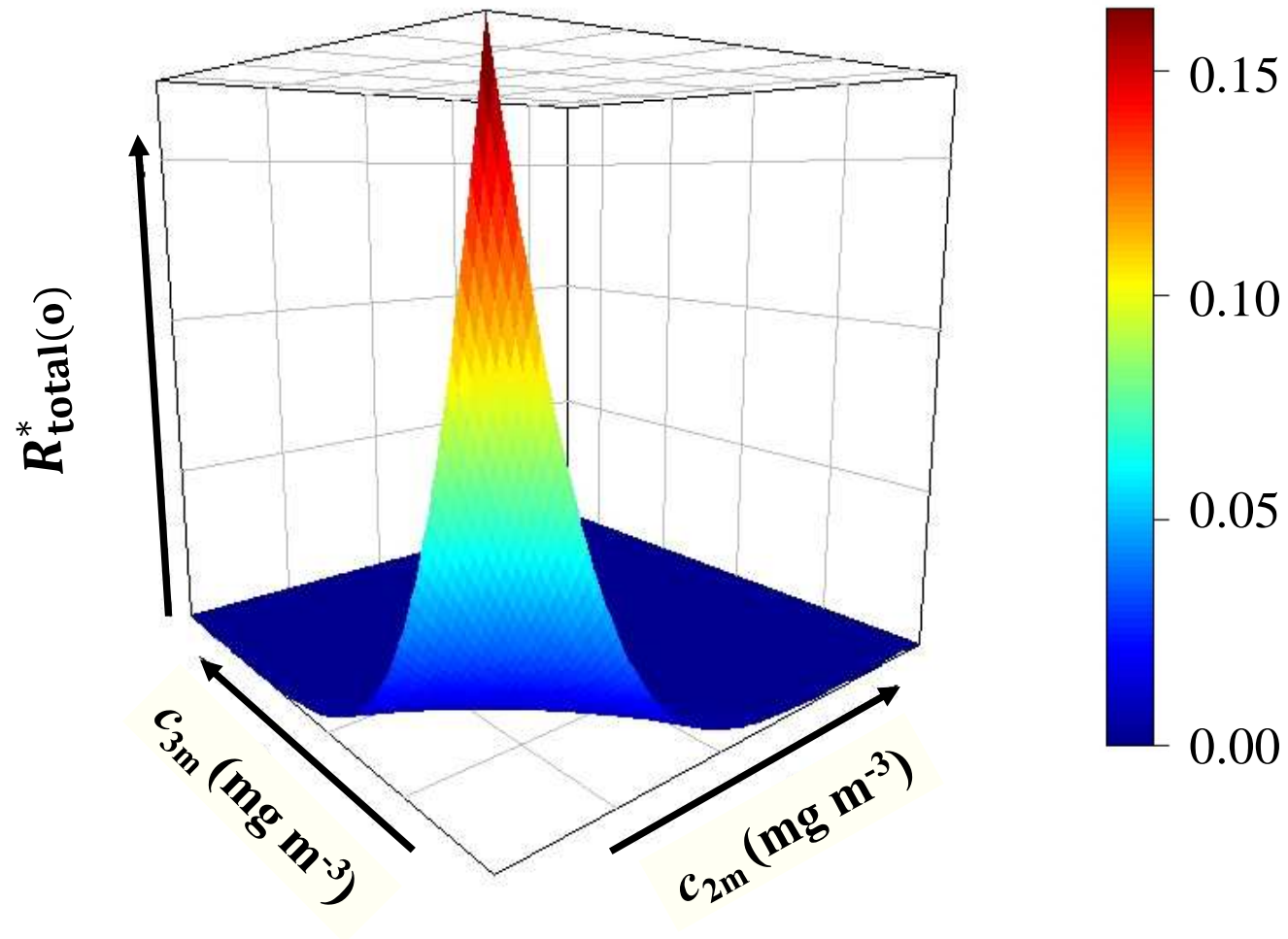
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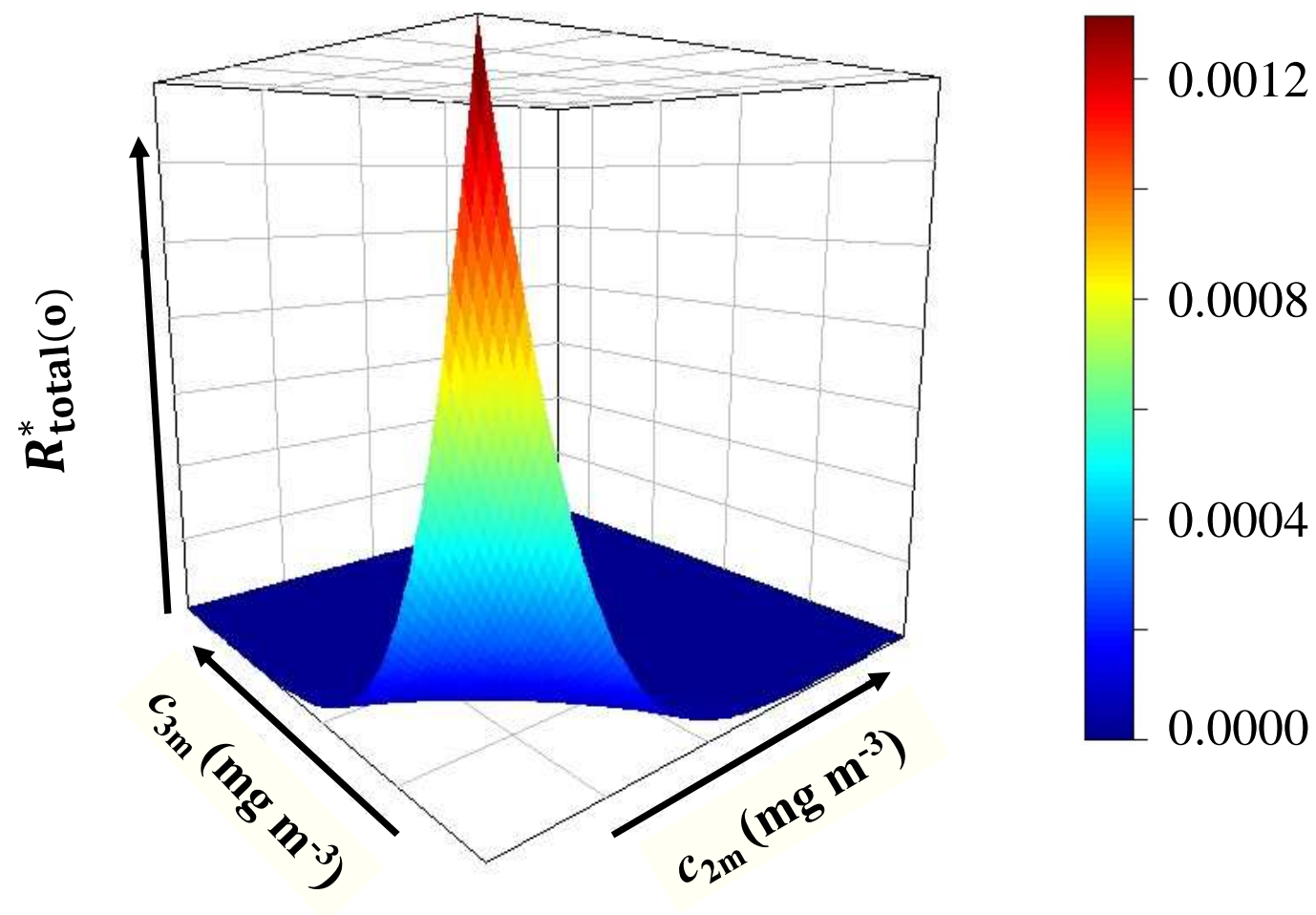
Figure captions

Fig. 1. Classification of the risks in conformity assessment of an environmental compartment due to measurement uncertainty. Specific risk refers to a specified compartment in a certain location at a certain time, whereas global risk – to the population of such compartments. Particular risk (specific R_{ci}^* or global R_{ci}) refers to i-th pollutant of the environmental compartment, $i = 1, 2, \ldots, n$, according to the JCGM Guide 106 (2012); and total risk (specific R_{total}^* or global R_{total}) – to the compartment as a whole. The total risk evaluation is the task of the IUPAC Project (2016), highlighted in the figure by an ellipse. These kinds of risks are relevant as for an underestimation of the pollutant concentration c_i, as for its overestimation, i.e. to the consumer' and producer's risks, respectively.

Fig. 2. Dependence of the total specific risks of underestimation $R_{total(u)}^*$ of TSPM concentration in air on the measurement results c_{im}. Fig. 2a is for a case when only the first quarry is active and the total risk $R_{total(u)}^*$ is equal to the particular risk $R_{c1(u)}^*$, shown by solid line 1. Dotted lines 3 and 2 point, as an example, a measured TSPM concentration $c_{1m} = 0.194$ mg m^{-3} and corresponding risk value $R_{c1(u)}^* = 0.211$. Fig. 2b is for a case when only the second and the third quarries are active. $R_{total(u)}^*$, presented as a color surface, is depending on both c_{2m} and c_{3m} in the range [0.010, 0.200] mg m^{-3}. The meaning of the color is the total risk value according to the color scale of the bar on the right side of the plot. Fig. 2c illustrates a case when all the three quarries are active and $R_{total(u)}^*$ - the color surface - is depending on c_{2m} and c_{3m} in the range [0.010, 0.200] mg m^{-3} as in Fig. 2b, but $c_{1m} = 0.194$ mg m^{-3} (indicated in Fig 2a by dotted lines).

Fig. 3. Dependence of the total specific risks of overestimation $R^*_{\text{total(o)}}$ of the TSPM concentration in air on the measurement results $c_{i\text{m}}$. Fig. 3a is for a case when only the first quarry is active and the total risk $R^*_{\text{total(o)}}$ is equal to the particular risk $R^*_{c1(\text{o})}$, shown by solid line 1, while dotted lines 3 and 2 point, as an example, a measured TSPM concentration $c_{1\text{m}} = 0.250$ mg m^{-3} and corresponding risk value $R^*_{c1(\text{o})} = 0.008$. Fig. 3b is for a case when only the second and the third quarries are active, as in Fig 2b, and the total risk $R^*_{\text{total(o)}}$ value is depending on both $c_{2\text{m}}$ and $c_{3\text{m}}$ in the range [0.210, 0.300] mg m^{-3}. Fig. 3c illustrates a case when all the three quarries are active simultaneously as in Fig. 2c, but $c_{1\text{m}} = 0.250$ mg m^{-3} (indicated in Fig 3a by dotted lines).

HIGHLIGHTS

- Evaluation of total risks of false decisions on conformity of an environmental compartment is developed.

- The total risks due to measurement uncertainty of concentrations of two or more pollutants are considered.

- As a case study, the total risks are evaluated at control of total suspended particulate matter (TSPM) concentration in air.

- The study concerns three independent stone quarries as pollutant sources.

- The total probabilities of under- and overestimation of TSPM concentration in air are calculated.