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Source apportionment and sensitivity analysis: two methodologies with two different purposes.

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Abstract

In this work are reviewed the existing methodologies for source apportionment and sensitivity analysis to identify key differences and stress their implicit limitations. The emphasis is laid on the differences between source "impacts" (sensitivity analysis) and "contributions" (source apportionment) obtained by using four different methodologies: brute force top-down, brute force bottom-up, tagged species and DDM. A simple theoretical example to compare these approaches is used highlighting differences and potential implications for policy. When the relationships between concentration and emissions are linear, impacts and contributions are equivalent concepts. In this case, source apportionment may be used for air quality planning purposes and vice versa, sensitivity analysis may be used for quantifying sources contributions. However, this study demonstrates that when the relationship between emissions and concentrations is non-linear, sensitivity approaches are not suitable to retrieve source contributions and source apportionment methods are not appropriate to evaluate the impact of abatement strategies. Moreover, when using sensitivity analysis for planning, it is important to note that, under non-linear circumstances, the calculated impacts will only provide information for the exact conditions (e.g. emission reduction share) that are simulated.

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1. Introduction

40
41 The concentration of a pollutant at a given location generally results
42 from direct emissions and from interactions in the atmosphere
43 among different emission precursors, emitted by a variety of
44 sources. For example, particulate matter (denoted here as PM)
45 results from the interaction and combination of 5 precursors (PPM,
46 NO_x, SO_2, NH_3, and VOC) which can be emitted by different
47 activity macro-sectors (e.g. residential, transport, industrial and
48 agriculture).
49
50 Abatement strategies aim at reducing the precursor's emission of the
51 different activity sector to reduce pollutant concentrations but these
52 strategies are challenging to design because of the complex
53 relationships that link precursors and pollutants.
54 Two different approaches are currently used to support air quality
55 decision makers: source apportionment and sensitivity analysis.
56
57 • Source apportionment quantifies the **<u>contribution</u>** of an
58   emission source (or precursor) to the concentration of one
59   pollutant at one given location.
60 • Sensitivity analysis estimates the **<u>impact</u>** on pollutant
61   concentration that results from a change of one or more emission
62   sources.
63
64 The main objective of this work is to review the existing
65 methodologies, identify key differences and stress their implicit
66 limitations. We will particularly focus on the differences between
67 concentration "impacts" (sensitivity) and "contributions" (source
68 apportionment) obtained with different methodologies. We make use
69 of a simple theoretical example to compare the approaches, highlight
70 differences and potential implications in terms of policy. In the
71 following sections, we analyze first how these methodologies work

3

72 in a simple linear case before generalizing it to more complex non-
73 linear situations.
74

2. Linear simplification and implications

76
77 Let's consider C a pollutant concentration at one location that is a
78 function of three variables (E_1, E_2 and E_3), i.e. the emissions of three
79 precursors or sources within a given domain: $C = C(E_1, E_2, E_3)$. For
80 a linear relationship between the function C and the three variables
81 E_1, E_2 and E_3, we can write:
82

$$C(E_1, E_2, E_3) = C(0,0,0) + P_1 E_1 + P_2 E_2 + P_3 E_3 \qquad (1)$$

83
84 where P_1, P_2 and P_3 are three constant coefficients.
85
86 On the other hand, the sensitivity of the concentration to a change of
87 a given emission source can be quantified via partial derivatives. For
88 expression (1) this gives:
89

$$\frac{\partial C}{\partial E_1} = P_1 \; ; \; \frac{\partial C}{\partial E_2} = P_2 \; ; \; \frac{\partial C}{\partial E_3} = P_3$$

90
91 In Clappier et al. (2017) the coefficients (P_1, P_2 and P_3) are referred
92 to "potencies". The Authors used this concept of "potencies" to
93 analyze the model response to emission changes in different
94 European countries.
95
96 The consequences of a linear relationship between concentration and
97 emission sources are twofold:
98 1) all higher order derivatives (order 2 and beyond) are null,
99 including those involving two or more emission sources (crossed
100 derivatives) as the impact of a change in one emission source is
101 independent from all others.
102 2) the first-order partial derivatives are constant and can therefore be
103 calculated with finite differencing, between any couple of emission

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104 levels, for example a base case (denoted BC) and a background
105 (denoted as 0).
106 The potency equations then read as:
107

$$P_1 = \frac{\Delta C_0^{BC_1}}{E_1^{BC}} \; ; \; P_2 = \frac{\Delta C_0^{BC_2}}{E_2^{BC}} \; ; P_3 = \frac{\Delta C_0^{BC_3}}{E_3^{BC}}$$

108

109 with $\Delta C_0^{BC_1} = C(E_1^{BC}, 0, 0) - C(0,0,0),$
110 $\Delta C_0^{BC_2} = C(0, E_2^{BC}, 0) - C(0,0,0)$
111 $\Delta C_0^{BC_3} = C(0,0, E_3^{BC}) - C(0,0,0)$

112

113 Together with "potencies", Clappier et al. also introduce the concept
114 of "potential", defined as the concentration change resulting from a
115 total reduction of the emissions (from BC to 0). The "potential" can
116 be calculated via relation (1) applied between the base-case and
117 background levels as:
118

$$\Delta C_0^{BC} = \Delta C_0^{BC_1} + \Delta C_0^{BC_2} + \Delta C_0^{BC_3} \qquad (2)$$

119

120 where $\Delta C_0^{BC} = C(E_1^{BC}, E_2^{BC}, E_3^{BC}) - C(0,0,0)$

121

122 Equation (2) can directly be used for source apportionment purpose,
123 with $\Delta C_0^{BC_1}$ the concentration change resulting from a total reduction
124 of the emission source (or precursor) E_1, reflecting the contribution
125 of E_1 to the base case concentration. Similarly $\Delta C_0^{BC_2}$ and $\Delta C_0^{BC_3}$ are
126 the contributions of E_2 and E_3. Equation (2) shows that, in the
127 linear case, the concentration change resulting from a simultaneous
128 reduction of all emission sources (ΔC_0^{BC}) is equal to the sum of the
129 emission sources contributions.

130

131 In the next sections, we will explore how this simple conclusion
132 changes when non-linear relationships are considered. In particular,
133 we will assess which implications (and limitations) these non-
134 linearities have in terms of source apportionment and sensitivity
135 analysis.

136

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3. Brute Force method

137

139 The "brute force" method consists in estimating the concentration
140 change by performing and subtracting two simulations, one with and
141 the second without a specific emission source to be analysed
142 (Blanchard, 1999; Yarwood et al., 2004).
143 In non-linear situations, the concentration change resulting from a
144 set of emission sources is not anymore equivalent to the sum of the
145 concentration changes resulting from emission sources changed
146 individually. In the following, we will refer to the work of Stein and
147 Alpert (1993) who proposed an approach to decompose an overall
148 impact into single (one emission source only) and combined
149 (multiple emission sources) impacts.

150
151
152   3.1. Bottom-up formulation
153
154
155 We consider here three emission precursors E_1, E_2 and E_3 that are
156 changing from a low (denoted as "L") to a high level (denoted as
157 "H"). In a bottom-up approach, the low emission level is chosen as
158 the reference. With these definitions and notations, the impact on
159 concentration resulting from a change of one only of the three
160 precursor's emissions can be written as follows:

161

$$\Delta C_L^{H_1} = C(E_1^H, E_2^L, E_3^L) - C(E_1^L, E_2^L, E_3^L)$$
$$\Delta C_L^{H_2} = C(E_1^L, E_2^H, E_3^L) - C(E_1^L, E_2^L, E_3^L)$$
$$\Delta C_L^{H_3} = C(E_1^L, E_2^L, E_3^H) - C(E_1^L, E_2^L, E_3^L)$$

162
163 While the impact on concentration resulting from the simultaneous
164 changes of two or three precursor's emissions would be written as:

165

$$\Delta C_L^{H_1,H_2} = C(E_1^H, E_2^H, E_3^L) - C(E_1^L, E_2^L, E_3^L)$$
$$\Delta C_L^{H_1,H_3} = C(E_1^H, E_2^L, E_3^H) - C(E_1^l, E_2^l, E_3^l)$$
$$\Delta C_L^{H_2,H_3} = C(E_1^L, E_2^H, E_3^H) - C(E_1^L, E_2^L, E_3^L)$$
$$\Delta C_L^{\overline{H}} = C(E_1^H, E_2^H, E_3^H) - C(E_1^L, E_2^L, E_3^L)$$

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166 Using a similar notation, the decomposition of Stein and Alpert
167 applied to 2 variables (E_1 and E_2) would read as:
168

$$\Delta C_{\bar{L}}^{H_1,H_2} = \Delta C_{\bar{L}}^{H_1} + \Delta C_{\bar{L}}^{H_2} + \hat{C}^{int} \qquad (3)$$

169
170
171 where $\Delta C_{\bar{L}}^{H_1}$ and $\Delta C_{\bar{L}}^{H_2}$ are the impacts induced by the change in
172 emission sources E_1 and E_2 taken independently and $\Delta C_{\bar{L}}^{H_1,H_2}$ the
173 impact induced from E_1 and E_2 taken simultaneously.
174
175 It is clear from (3) that the impact of a simultaneous change of two
176 emission sources is not equivalent to the sum of the individual
177 impacts, as highlighted by the additional term $\hat{C}^{int}$. This term that
178 quantifies the interaction between the two emission sources can be
179 calculated using (3) as:
180

$$\hat{C}^{int} = \hat{C}_{\bar{L}}^{H_1,H_2} = \Delta C_{\bar{L}}^{H_1,H_2} - \Delta C_{\bar{L}}^{H_1} - \Delta C_{\bar{L}}^{H_2} \qquad (4)$$

181
182 The Stein and Alpert formulation can similarly be applied with 3
183 emission sources:
184

$$\Delta C_{\bar{L}}^{\bar{\bar{H}}} = \Delta C_{\bar{L}}^{H_1} + \Delta C_{\bar{L}}^{H_2} + \Delta C_{\bar{L}}^{H_3} + \hat{C}^{int} \qquad (5)$$

185
186 Where $\Delta C_{\bar{L}}^{H_1}$, $\Delta C_{\bar{L}}^{H_2}$ and $\Delta C_{\bar{L}}^{H_3}$ are the impact on concentration
187 resulting from single emission changes in the sources and
188

$$\hat{C}^{int} = \hat{C}_{\bar{L}}^{H_1,H_2} + \hat{C}_{\bar{L}}^{H_1,H_3} + \hat{C}_{\bar{L}}^{H_2,H_3} + \hat{C}_{\bar{L}}^{H_1,H_2,H_3} \qquad (6)$$

189
190 where $\hat{C}_{\bar{L}}^{H_1,H_2}$, $\hat{C}_{\bar{L}}^{H_1,H_3}$ and $\hat{C}_{\bar{L}}^{H_2,H_3}$ are the double interaction terms that
191 can be further decomposed via equation (4). $\hat{C}_{\bar{L}}^{H_1,H_2,H_3}$ is the triple
192 interaction term (between E_1, E_2, E_3) which can be decomposed by
193 combining (5) and (6) as:
194

7

$$\hat{C}_{\bar{L}}^{H_1,H_2,H_3} = \Delta C_{\bar{L}}^{\bar{H}} - \Delta C_{\bar{L}}^{H_1} - \Delta C_{\bar{L}}^{H_2} - \Delta C_{\bar{L}}^{H_3}$$

$$\Delta C_{\bar{L}}^{H_1,H_2} - \Delta C_{\bar{L}}^{H_1,H_3} - \Delta C_{\bar{L}}^{H_2,H_3}$$

195

196   3.2. Top-down formulation

197

198   In a top-down formulation, the highest emission level is chosen as
199   reference. The Stein Alpert formulation for three precursors can then
200   be expressed similarly to the bottom-up formulation as:

201

$$\Delta C_{\bar{L}}^{\bar{H}} = \Delta C_{L_1}^{\bar{H}} + \Delta C_{L_2}^{\bar{H}} + \Delta C_{L_3}^{\bar{H}} + \hat{C}^{int} \qquad (7)$$

202

203   where $\Delta C_{L_1}^{\bar{H}}$, $\Delta C_{L_2}^{\bar{H}}$ and $\Delta C_{L_3}^{\bar{H}}$ are the impacts on concentration induced
204   by reducing E_1 , E_2 and E_3 independently whereas $\hat{C}^{int}$ is the
205   interaction term which itself can be decomposed into a series of
206   double interactions and a triple interaction terms:

207

$$\hat{C}^{int} = \hat{C}_{L_1,L_2}^{\bar{H}} + \hat{C}_{L_1,L_3}^{\bar{H}} + \hat{C}_{L_2,L_3}^{\bar{H}} + \hat{C}_{L_1,L_2,L_3}^{\bar{H}} \qquad (8)$$

208

209   It is important to stress that the top-down single impacts are not
210   equivalent to their bottom-up counterparts. The relation between
211   these bottom-up and top-down impacts can be expressed as (here for
212   the case of E_3):

213

$$\Delta C_{L_3}^{H} = C(E_1^H, E_2^H, E_3^H) - C(E_1^H, E_2^H, E_3^L)$$

214

215

$$\Delta C_{L_3}^{H} = C(E_1^H, E_2^H, E_3^H) - C(E_1^L, E_2^L, E_3^H)$$
$$- [C(E_1^H, E_2^H, E_3^L) - C(E_1^L, E_2^L, E_3^L)]$$

216

$$\Delta C_{L_3}^{\bar{H}} = \Delta C_{\bar{L}}^{H} - \Delta C_{\bar{L}}^{H_1 H_2} \qquad (9)$$

217

218   Using equations (3), (4), (5) and (6), equation (9) can be re-
219   expressed as:

220

8

$$\Delta C_{L_3}^{\bar{H}} = \Delta C_{\bar{L}}^{H_3} + \hat{C}_{\bar{L}}^{H_1,H_3} + \hat{C}_{\bar{L}}^{H_2,H_3} + \hat{C}_{\bar{L}}^{H_1,H_2,H_3} \qquad (10)$$

221
222 In other words, the top-down impact on concentration of an emission
223 source (obtained by switching off the emission source while all
224 others remain unchanged) is not equivalent to its bottom-up
225 counterpart, (obtained by switching on the emission source while all
226 others are switched off). Relation (10) indeed clearly shows that
227 additional interaction terms need to be considered. The implications
228 resulting from these differences are highlighted in Section 5 in
229 which some theoretical examples are described.
230

231 **4. Source apportionment and sensitivity analysis**

232
233
234   4.1. Tagged species techniques

235
236 Equation (2) shows that, when the relationship between
237 concentration and several emission sources is linear, the contribution
238 of a specific source (source apportionment) can be computed as the
239 impact on concentration obtained by a full reduction of this source
240 (sensitivity). Moreover, the sum of the impacts on concentration
241 obtained by reduction of the single sources ($\Delta C_0^{BC_1} + \Delta C_0^{BC_2} + \Delta C_0^{BC_3}$) is
242 equivalent to the impact on concentration resulting from a
243 simultaneous abatement of all sources (ΔC_0^{BC}). In such a case, the
244 concentration impacts are equal to source contributions and source
245 apportionment and sensitivity analysis lead to similar results. This is
246 not the case, however, when the relationship between concentrations
247 and emissions is nonlinear. In their approach, Stein and Alpert
248 express the difference between the impact caused by a simultaneous
249 abatement and the sum of the impacts caused by individual
250 abatement as interactions among the different sources. The Stein and
251 Alpert formulation applied between the base-case and background
252 levels is very close to equation (2) but with an additional term that
253 accounts for interactions:
254

$$\Delta C_0^{BC} = \Delta C_0^{BC_1} + \Delta C_0^{BC_2} + \Delta C_0^{BC_3} + \hat{C}^{int}$$

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255
256 Because the interaction terms cannot not be attributed to a single
257 emission source as they represent the interaction between two and
258 more emission sources, the Stein and Alpert methodology does not
259 allow identifying the full contribution of each individual source. It
260 cannot therefore be used for source apportionment purpose, unless
261 the interaction terms are negligible as in the linear case.
262 Unlike the Stein and Alpert methodology, the tagged species
263 methodology is designed for source apportionment purposes. This
264 methodology tags each precursor and quantifies its contribution (in
265 terms of mass) to the pollutant concentration.
266 Tagged algorithms are implemented in several chemical transport
267 model systems (Yarwood et al., 2004; Wagstrom et al., 2008,
268 ENVIRON, 2014; Bhave et al., 2007; Wang et al., 2009;
269 Kranenburg et al., 2013).
270
271 In tagging approaches, the effect of the full reduction of all sources
272 is directly expressed as the sum of the source contributions:
273

$$\Delta C_0^{BC} = \delta C_1 + \delta C_2 + \delta C_3$$

274
275 where δC_1, δC_2, δC_3 are the contributions of sources E_1, E_2 and E_3
276 resulting from the tagged species approach resolution.
277
278 Tagging methodologies split the interaction terms into fractions and
279 attribute these fractions to the source contributions, on the basis of
280 mass weighting factors.
281

$$\delta C_1 = \Delta C_0^{BC_1} + \alpha \hat{C}^{int}$$

282
283
284 Because the tagged species approach mixes interaction terms and
285 single concentration impacts into sources contributions, it is not
286 suitable to estimate the effect of emission reduction when non-
287 linearities are present (Burr and Zhang 2011a, 2011b). Indeed these
288 two types of terms may react in very different ways to emission
289 reductions. This fact is detailed in the examples provided below.

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290 On the other hand, the strength of this method is that it allows for a
291 direct comparison of the source contributions with measurements (or
292 measurement based methods like receptor models).
293
294
295   4.2. DDM
296
297 The decoupled direct method (DDM) is designed to estimate
298 sensitivities to emission changes (Dunker et al., 1984; Dunker et al.,
299 2002). It aims to compute only the first order derivatives (which
300 correspond to the potencies mentioned in paragraph 2).
301

$$\frac{\partial C}{\partial E_1} \ ; \ \frac{\partial C}{\partial E_2} \ ; \ \frac{\partial C}{\partial E_3}$$

302
303 The Taylor formula is applied at first order to calculate the
304 concentration change between two emission levels (denoted H and
305 L).
306

$$\Delta C_L^{\bar{H}} = \Delta E_1 \left.\frac{\partial C}{\partial E_1}\right|_H + \Delta E_2 \left.\frac{\partial C}{\partial E_2}\right|_H + \Delta E_3 \left.\frac{\partial C}{\partial E_3}\right|_H$$

307
308   with $\Delta E_1 = E_1^H - E_1^L, \Delta E_2 = E_2^H - E_2^L, \Delta E_3 = E_3^H - E_3^L$
309
310 In the linear case, the first order derivatives are constant and the first
311 order approximation of the Taylor formula gives the exact
312 expression of the impact on concentration of an emission change
313 between H and L. When the emission-concentration relationship is
314 nonlinear, the first derivatives are not constants. The first order
315 Taylor formula cannot take into account the nonlinear effects. It is a
316 linear approximation based on derivatives computed at a given
317 emission reference level (level H in our example). The estimation of
318 the impact on concentration of an emission change between H and L
319 is accurate enough if level L is close enough to level H.
320
321 The HDDM method (Hakami et al., 2003) aim to increase the
322 accuracy of the DDM method by computing second order
323 derivatives.

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324   DDM (and HDDM) gives similar information to the Stein Alpert
325   formulation applied with the Brute force top-down approach
326   (because the reference level is H). For the same reason than for the
327   Stein Alpert approach, these two methods are suitable for source
328   apportionment purpose only if the relation between concentration
329   and emission is close to linearity.
330   DDM (and HDDM) approximates the impact on concentration from
331   an emission change between the two level H and L, using
332   derivatives computed at level H. This impact is accurate enough if
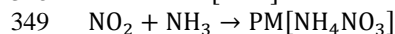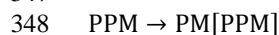333   the level L is close enough to the reference level H.
334
335

336   **5. Example**

337
338   In this section, examples are designed to illustrate the differences in
339   terms of contribution and impact estimates when the approaches
340   discussed previously are used. In these examples, we focus on the
341   formation of particulate matter (PM) in the atmosphere and only
342   consider three formation processes: direct emissions (primary PM
343   denoted as PPM); formation through reactions with nitrogen oxides
344   (NO_2) and ammonia (NH_3) and formation through reactions with
345   sulphur oxide (SO_2) and NH_3. These reactions pathways are
346   summarised by the following system of reactions:

347
348    $PPM \rightarrow PM[PPM]$
349    $NO_2 + NH_3 \rightarrow PM[NH_4NO_3]$
350    $SO_2 + 2\,NH_3 \rightarrow PM[(NH_4)_2SO_4]$
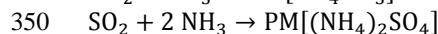
351
352   This system is further simplified by assuming that all reactions have
353   comparable kinetics (reaction speed) and have reached their
354   equilibrium. From these three reactions, 1 PM mole can be produced
355   by 1 PPM mole, by the combination of 1 NH_3 and 1 NO_2 moles or
356   by the combination of 1 SO_2 and 2 NH_3 moles.

357
358   We also limit our examples to emissions from three activity sectors.
359   The residential sector (R) only emits PPM and NO_2, the agricultural
360   sector (A) only emits NH_3 and the industrial sector (I) only emits
361   PPM and SO_2 (Figure 1). We assume for convenience that no

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362   background pollution is present (i.e. there is no PM when all
363   emissions are zero).
364
365   **5.1. Non-limited regime**
366
367   In this first example, the quantity of precursors (in terms of mass) is
368   large enough to feed all reactions. The agricultural sector emits 150
369   NH_3 moles which can react with 50 NO_2 moles emitted by the
370   residential sector and 50 SO_2 moles emitted by industrial sector. 100
371   PPM moles are emitted by the residential sector as well by the
372   industrial sector.
373
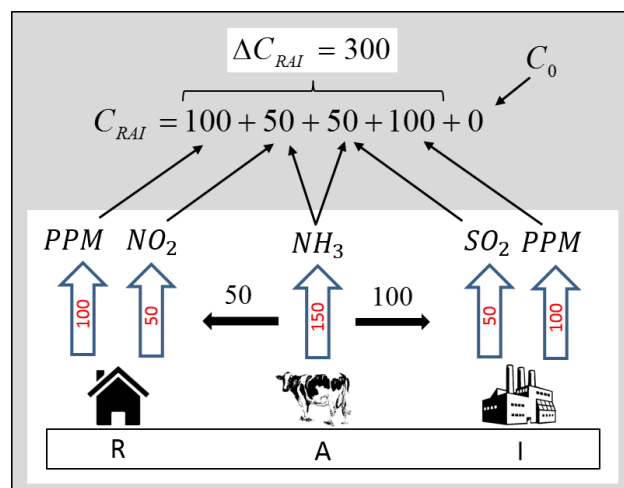
374
375   Figure 1. Example of PPM, NO_2, SO_2 and NH_3 emissions released
376   by three activity sectors: residential (R), agricultural (A) and
377   industrial (I). For convenience, no units are associated to emissions
378   and concentrations
379
380   Let's first calculate the PM concentration produced with and without
381   each of the sources:
382

383   No source: C_0 is the PM concentration obtained when all emissions
384   are set to zero. Since we assumed a zero background pollution,
385   $C_0 = 0$.
386

387   One source only: C_R (resp. C_A and C_I) is the PM concentration
388   reached when only the residential (resp. agricultural and industrial)
389   sector releases emissions:
390   - $C_R = 100$ produced by PPM emissions (NO_2 emissions do not
391   produce PM as no NH_3 is available).
392   - $C_A = 0$ because NO_2 and SO_2 are not available to react with
393   NH_3.
394   - $C_I = 100$ produced by the PPM emissions (SO_2 emissions do not
395   produce PM as no NH_3 is available).
396

397   Two sources: C_{RA} C_{RI}, and C_{AI} are the concentrations obtained when
398   two (out of three) activity sectors release their emissions
399   simultaneously (the RA subscripts correspond to residential and
400   agriculture, RI to residential and industrial, AI to agriculture and
401   industrial):
402   - $C_{RA} = 150$: 100 produced by PPM emissions from the
403   residential sector and 50 produced by the 50 NO_2 released by the
404   residential sector reacting with the 50 NH_3 emitted by agriculture
405   (100 NH_3 moles remain unused).
406   - $C_{RI} = 200$: 100 produced by PPM emissions from the residential
407   sector and 100 produced by PPM emissions from the industrial
408   sector.
409   - $C_{AI} = 150$: 100 produced by PPM industrial emissions and 50
410   from the combination of 50 SO_2 (industry) and 100 NH_3
411   (agriculture).
412

413   Three sources: C_{RAI} is the concentrations obtained when all
414   emissions are released simultaneously.
415   - $C_{RAI} = 300$: 200 from PPM (residential and industry), 50 from
416   reaction between NO_2 and NH_3 and 50 from reaction between SO_2
417   and NH_3.
418

419   **Brute-force Bottom-up (BF-BU) method**
420

421   The contribution of each activity sector is calculated as the
422   concentration change resulting from a 100% emission increase from
423   the lowest emission level (previously denoted "L" or background) to
424   the highest level (denoted as "H", the base case C_{RAI} obtained with
425   all emissions)

426

427   In a bottom-up approach, the concentration associated with the
428   lowest emission level is considered as the reference. Concentration
429   impacts are then computed by difference between any situation (e.g.
430   one, two or three sources present) and this reference.

431

432   - with one source: $\Delta C_R^{BU} = C_R - C_0 = 100$
433      $\Delta C_A^{BU} = C_A - C_0 = 0$
434      $\Delta C_I^{BU} = C_I - C_0 = 100$

435

436   - with two sources: $\Delta C_{RA}^{BU} = C_{RA} - C_0 = 150$
437      $\Delta C_{RI}^{BU} = C_{RI} - C_0 = 200$
438      $\Delta C_{AI}^{BU} = C_{AI} - C_0 = 150$

439

440   - with three sources: $\Delta C_{RAI} = C_{RAI} - C_0 = 300$

441

442   To calculate the interaction terms, we use the Stein-Alpert
443   formulation using Eq. (5) and Eq. (6):

444

$$\Delta C_{RAI} = \Delta C_R^{BU} + \Delta C_A^{BU} + \Delta C_I^{BU} + \hat{C}_{RA}^{BU} + \hat{C}_{RI}^{BU} + \hat{C}_{AI}^{BU} + \hat{C}_{RAI}^{BU}$$

445

446   from which the interaction terms are obtained by application of (4)
447   and (6):

448

$$\hat{C}_{RA}^{BU} = \Delta C_{RA}^{BU} - \Delta C_R^{BU} - \Delta C_A^{BU} = 50$$
$$\hat{C}_{RI}^{BU} = \Delta C_{RI}^{BU} - \Delta C_R^{BU} - \Delta C_I^{BU} = 0$$
$$\hat{C}_{AI}^{BU} = \Delta C_{AI}^{BU} - \Delta C_A^{BU} - \Delta C_I^{BU} = 50$$
$$\hat{C}_{RAI}^{BU} = \Delta C_{RAI} - \Delta C_R^{BU} - \Delta C_A^{BU} - \Delta C_I^{BU} - \hat{C}_{RA}^{BU} - \hat{C}_{RI}^{BU} - \hat{C}_{AI}^{BU} = 0$$

449

450   As can be seen from this example, the system behaves non-linearly
451   and the interaction terms (e.g. $\hat{C}_{RA}^{BU}$) are not-zero. Moreover the sum
452   of the individual impacts ($\Delta C_R^{BU} + \Delta C_A^{BU} + \Delta C_I^{BU} = 200$)
453   underestimates the overall impact ($\Delta C_{RAI} = 300$). These results are
454   graphically represented in Figure 2 (third column).

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455
456
457 **Brute-force Top-down (BF-TD) method**
458
459 In a BF-TD approach, the higher emission level (base case, C_{RAI}) is
460 the reference and the impact of each activity sector is calculated as
461 the concentration change resulting from a 100% emission decrease
462 (of one, two or three sources) from this reference to the background
463 level.
464
465 - with one source: When all emissions from one sector are
466 reduced (e.g. residential), the other two sector remain active
467 (agricultural and industry). In this case, the Top-down impact is
468 the difference between the base case concentration and the
469 concentration resulting from the agricultural and industrial
470 emissions only. A similar reasoning can be made for all sectors:
471
472 $$\Delta C_R^{TD} = C_{RAI} - C_{AI} = 150$$
473 $$\Delta C_A^{TD} = C_{RAI} - C_{RI} = 100$$
474 $$\Delta C_I^{TD} = C_{RAI} - C_{RA} = 150$$
475
476 - with two sources: The Top-down impact due to a full reduction
477 of two sectors (e.g. residential and agriculture) is similarly
478 computed as the difference between the base case concentration
479 and the concentration resulting from the remaining sector
480 (industry).
481
482 $$\Delta C_{RA}^{TD} = C_{RAI} - C_I = 200$$
483 $$\Delta C_{RI}^{TD} = C_{RAI} - C_A = 300$$
484 $$\Delta C_{AI}^{TD} = C_{RAI} - C_R = 200$$
485
486
487 - with three sources: The impact resulting from the simultaneous
488 reduction of all three sources is similar in the Top-down and
489 Bottom-up approaches:
490
491 $$\Delta C_{RAI} = C_{RAI} - C_0 = 300$$
492

493 The interaction terms can be obtained in a similar way to the
494 bottom-up approach by using the Stein and Alpert formulation for
495 ΔC_{RAI}:
496

$$\Delta C_{RAI} = \Delta C_R^{TD} + \Delta C_A^{TD} + \Delta C_I^{TD} + \hat{C}_{RA}^{TD} + \hat{C}_{RI}^{TD} + \hat{C}_{AI}^{TD} + \hat{C}_{RAI}^{TD}$$

497
498 The interaction terms are given by:
499

$$\hat{C}_{RA}^{TD} = \Delta C_{RA}^{TD} - \Delta C_R^{TD} - \Delta C_A^{TD} = -50$$
$$\hat{C}_{RI}^{TD} = \Delta C_{RI}^{TD} - \Delta C_R^{TD} - \Delta C_I^{TD} = 0$$
$$\hat{C}_{AI}^{TD} = \Delta C_{AI}^{TD} - \Delta C_A^{TD} - \Delta C_I^{TD} = -50$$
$$\hat{C}_{RAI}^{TD} = \Delta C_{RAI} - \Delta C_R^{TD} - \Delta C_A^{TD} - \Delta C_I^{TD} - \hat{C}_{RA}^{TD} - \hat{C}_{RI}^{TD} - \hat{C}_{AI}^{TD} = 0$$

500
501 With this approach, a non-linear behavior is also observed and
502 interaction terms are not zero. It is also interesting to note that the
503 triple interaction term ($\hat{C}_{RAI}^{TD}$) is null. The sum of the individual
504 impacts ($\Delta C_R^{TD} + \Delta C_A^{TD} + \Delta C_I^{TD} = 400$) overestimates the overall
505 impact ($\Delta C_{RAI} = 300$). We further discuss these aspects at the end of
506 this section. These results are graphically represented in Figure 2
507 (fourth and fifth columns).
508
509
510 **Tagged species approach**
511
512 Compared to Brute-force, the tagged species approach calculates the
513 share of each source to the overall concentration change. These
514 shares are referred to as contributions and have the main property
515 that the sum of the individual contributions is equal to the overall
516 concentration impact, by definition, i.e.:
517

$$\Delta C_{RAI} = \delta C_R^{TAG} + \delta C_A^{TAG} + \delta C_I^{TAG}$$

518
519
520 The sector contributions are computed by tracking the mass of their
521 emitted species contributing to PM formation (in our example:
522 $PM[PPM]$, $PM[NH_4NO_3]$ and $PM[(NH_4)_2SO_4]$)
523

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524   - PM[PPM] is formed from PPM. The 100 moles from the
525   residential sector lead to 100 moles of PM. The same applies to
526   the 100 moles from industry.
527
528   - PM[NH_4NO_3] is formed by combination of NH_3 and NO_2. The
529   share between these two contributions is obtained by application
530   of stoichiometric molar mass ratios:
531

$$a_1 = \frac{[NO_3]_m}{[NO_3]_m + [NH_4]_m} = 0.78$$

532
533   In our example, 50 moles of PM[NH_4NO_3] are formed by
534   combination of NO_2 (50 moles) from the residential sector and
535   NH_3 (50 moles) from agriculture. The contribution attributed to
536   NO_2 is $50 \times a_1$ whereas the contribution attributed to NH_3 is
537     $50 \times (1 - a_1)$.
538
539
540   - PM[$(NH_4)_2SO_4$] is formed by combination of NH_3 and SO_2. The
541   following stoichiometric mass ratio is used:
542

$$a_2 = \frac{[SO_4]_m}{[SO_4]_m + 2[NH_4]_m} = 0.73$$

543
544   The contribution attributed to SO_2 is $50 \times a_2$ whereas the
545   contribution attributed to NH_3 is $50 \times (1 - a_2)$.
546
547   The contribution of each sector is then obtained as the sum of their
548   precursor contribution shares as follows:
549
550   $\delta C_R^{TAG} = 100 + 50 \times a_1 = 138.7$
      $\delta C_A^{TAG} = 50 \times (1 - a_1) + 50 \times (1 - a_2) = 24.9$
      $\delta C_I^{TAG} = 100 + 50 \times a_2 = 136.4$
551
552   By definition the sum of the contributions ($\delta C_R^{TAG} + \delta C_A^{TAG} +$
553   $\delta C_I^{TAG} = 300$) is exactly equal to the overall concentration impact
554   ($\Delta C_{RAI} = 300$).
555

556 Note that a decomposition of the non-linear interaction terms
557 obtained in the top-down or bottom-up approach, using the above
558 stoichiometric ratios would lead to similar results than the tagged
559 approach. These results are graphically represented in Figure 2
560 (second column).

561

562

563 **DDM**

564

565 In this methodology, delta concentrations and interaction terms are
566 estimated with first order partial derivatives computed from the
567 highest emission level (base case in our example). Being a
568 sensitivity approach using level H as reference, DDM shows clear
569 analogies with the BF-TD.

570

571 $$\left.\frac{\partial C}{\partial \alpha_R}\right|_{TD} = 150 \qquad \left.\frac{\partial C}{\partial \alpha_A}\right|_{TD} = 100 \qquad \left.\frac{\partial C}{\partial \alpha_I}\right|_{TD} = 150$$

572

573

574 where α_R , α_A and α_I are percentage emission changes from the base
575 case for the residential, agricultural and industrial sectors.
576 The first-order derivatives are evaluated using finite differencing
577 between the base case and a level characterised by emissions that are
578 10% lower for each activity sector.
579 The concentration changes resulting from a 100% emission
580 reduction (i.e. between the base case and the zero emission case)
581 could be estimated by setting α_R, α_A and α_I to unity.

582

$$\Delta C_R^{HDDM} = \left.\frac{\partial C}{\partial \alpha_R}\right|_{TD} = 150$$

$$\Delta C_A^{HDDM} = \left.\frac{\partial C}{\partial \alpha_A}\right|_{TD} = 100$$

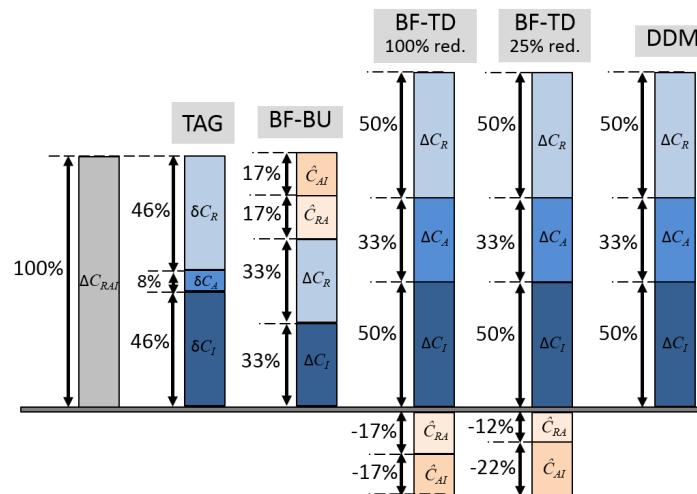$$\Delta C_I^{HDDM} = \left.\frac{\partial C}{\partial \alpha_I}\right|_{TD} = 150$$

583

584

585 We see from this last example that both the total PM and the
586 contribution of the sources are then comparable with those obtained

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587 with the BF-TD method. Their interpretation is similar (Figure 2,
588 sixth column).
589

590
591 Figure 2. Schematic representation of the allocation of PM to its
592 sources in the non-limited example. The expected total PM is
593 displayed in the grey bar on the left.
594
595 In the linear case (second paragraph) we have seen that a single
596 source contribution can be computed as the impact resulting from a
597 full reduction of this source. However, source contributions and
598 concentration impacts should not be confused as they are different in
599 most situations. The example presented in this paragraph illustrates
600 this clearly for a non-linear system. Indeed the contributions of the
601 single sources computed by the tagged species approach ($\delta C_R^{TAG} =$
602 138 ; $\delta C_A^{TAG} = 24$; $\delta C_I^{TAG} = 136$) differ from the concentration
603 impacts resulting from a total abatement of these single sources
604 computed by the BF-TD ($\Delta C_R^{TD} = 150$; $\Delta C_A^{TD} = 100$; $\Delta C_I^{TD} = 150$)
605 method. Moreover, the sum of the concentration impacts obtained
606 with either the BF-TD approach for single sources does not equal the
607 total concentration impact ($\Delta C_{RAI} = 300$). This is also valid for any
608 selection of two sectors ($\Delta C_R^{TD} + \Delta C_A^{TD} = 250 \neq \Delta C_{RA}^{TD} = 200$). Note

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20

609   that similarly to BF-TD, the concentration impacts computed as
610   increases from the background (BF-BU) show the same behaviour
611   ($\Delta C_R^{BU} + \Delta C_A^{BU} = 100 \neq \Delta C_{RA}^{BU} = 150$).
612
613   Figure 3 shows that the impact on concentration is proportional to
614   the emission reduction indicating that the relationship between
615   emission and concentration changes is linear. However, this example
616   also illustrates the fact that linearity encompasses two aspects: 1)
617   the interaction terms are zero ($\hat{C}^{int} = 0$); 2) the ratios between
618   concentration change and emission changes ($\Delta C / \Delta E$) remain
619   constant, regardless of the calculation bounds (denoted "H" and "L"
620   in Section 4). In the current example the ratios $\Delta C / \Delta E$ are constant
621   (linear trend of ΔC on Figure 3) but the relationship between
622   concentration and emission is not linear because of the non-zero
623   interaction terms (not shown) ($\hat{C}_{RA}^{TD} = -50$ and $\hat{C}_{AI}^{TD} = -50$).
624   However, even with zero interaction terms, we can still observe a
625   non-linear behaviour with the emission reduction percentage. The
626   evaluation of linearity therefore requires two tests: one to quantify
627   the interaction terms and the second to assess the deviation from a
628   linear trend with respect to the emission reduction percentage.
629
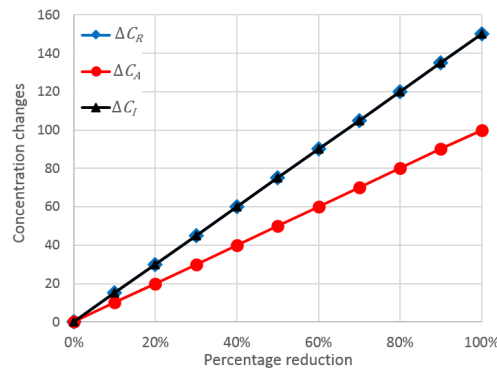
630
631   Figure 3: Evolution of the concentration changes resulting from
632   different percentage of source abatement (Top down approach) for
633   the three sectors (Residential, Agricultural and Industrial).
634

635
636
637   **5.2. Limited regime**

638
639   This example is similar to the previous, except that the emissions of
640   NH_3 are reduced from 150 to 100 moles.
641   When all sources release emissions, the 100 moles of NH_3 are
642   shared into 100/3=33.3 moles which react with NO_2 to form 33.3
643   moles of $PM[NH_4NO_3]$ and $100\times2/3$=66.6 moles which react with
644   SO_2 to give 33.3 moles of $PM[(NH_4)_2SO_4]$. Because the mass of
645   NH_3 is not enough to react with all the NO_2 and SO_2 mass, 16.7
646   moles of NO_2 and 16.7 moles of SO_2 remain unused.
647   Note that when the agricultural source is active with one only of the
648   two other sources (residential or industrial) the NH_3 100 moles are
649   then sufficient to consume all the NO_2 or SO_2 and lead to 50 moles
650   of PM in either case.
651
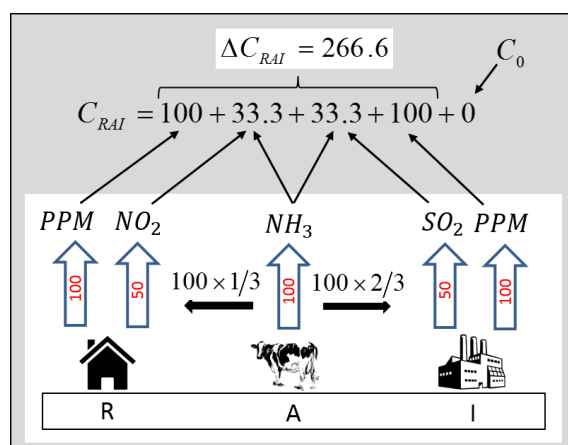
652
653   Figure 3. Example with three sources in an ammonia-limited regime.
654   The mass emitted by each source is expressed in moles.

655
656   The PM concentrations obtained when one or two sources are active
657   are similar to the previous example:
658

659    $C_0 = 0 \; ; \; C_R = 100 \; ; \; C_A = 0 \; ; \; C_I = 100$
660    $C_{RA} = 150 \; ; \; C_{RI} = 200 \; ; \; C_{AI} = 150$
661
662   But the result differ when all sources are active: $C_{RAI} = 266.6$ (200
663   from PPM (residential industry), 33.3 from reaction between NO_2
664   and NH_3 and 33.3 from reaction between SO_2 and NH_3).
665
666   **Bottom-up brute-force method (BF-BU)**
667
668   The BF-BU approach computes all concentration impacts from the
669   background concentration (C_0). The Stein-Alpert terms are similar to
670   the non-limited case, excepted for ΔC_{RAI} and $\hat{C}_{RAI}$:
671
672        $\Delta C_R^{BU} = 100$ $\hat{C}_{RA}^{BU} = 50$
673        $\Delta C_A^{BU} = 0$ $\hat{C}_{RI}^{BU} = 0$
674        $\Delta C_I^{BU} = 100$ $\hat{C}_{AI}^{BU} = 50$
675        $\Delta C_{RAI} = 266.6$ $\hat{C}_{RAI} = -33.3$
676
677   The limiting effect of NH_3 appears only in the negative triple
678   interaction term ($\hat{C}_{RAI}$).
679
680   **Top-down brute-force method (BF-TD)**
681
682   The Top-down approach uses the base case (C_{RAI}) concentration as
683   reference to compute the concentration impact. In this case, all
684   Stein-Alpert terms are different from the non-limited regime:
685
686        $\Delta C_R^{TD} = 116.6$ $\hat{C}_{RA}^{TD} = -16.6$
687        $\Delta C_A^{TD} = 66.6$ $\hat{C}_{RI}^{TD} = 33.3$
688        $\Delta C_I^{TD} = 116.6$ $\hat{C}_{AI}^{TD} = -16.6$
689        $\Delta C_{RAI} = 266.6$ $\hat{C}_{RAI} = -33.3$
690
691
692   **Tagged approach**
693
694   The contribution of each source is computed similarly to the non-
695   limited regime. The productions of 33.3 moles of $PM[NH_4NO_3]$ and

696   33.3 moles of PM[$(NH_4)_2SO_4$] are split among the different sectors
697   using the stoichiometric coefficients a_1 and a_2:

698

699   $\delta C_R^{TAG} = 100 + 33.3 * a_1 = 125.8$
      $\delta C_A^{TAG} = 33.3 * (1 - a_1) + 33.3 * (1 - a_2) = 16.6$
      $\delta C_I^{TAG} = 100 + 33.3 * a_2 = 124.2$

700
701
702   **DDM**

703

704   As shown below, DDM only considers first derivatives, which are
705   not suitable to estimate higher order interaction terms. The
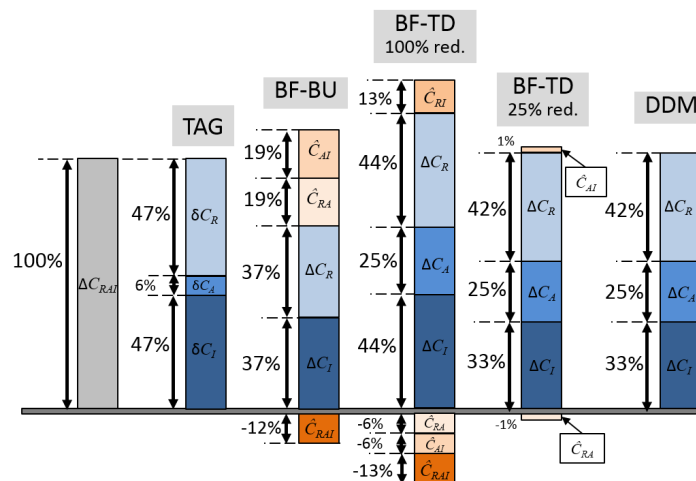706   calculation of the first order derivatives in this example gives:

707

$$\Delta C_R^{HDDM} = \left.\frac{\partial C}{\partial \alpha_R}\right|_{TD} = 111.5$$

$$\Delta C_A^{HDDM} = \left.\frac{\partial C}{\partial \alpha_A}\right|_{TD} = 66.7$$

$$\Delta C_R^{HDDM} = \left.\frac{\partial C}{\partial \alpha_I}\right|_{TD} = 88.1$$

708

709

710 Figure 4. Schematic representation of the allocation of PM to its
711 sources in the ammonia-limited example. The expected total PM is
712 displayed in the grey bar on the left.
713
714 The main difference with respect to the non-limited regime is the
715 appearance of a triple interaction term that will also lead to
716 differences between the BF-TD and the DDM approaches, given the
717 fact that the latter only accounts for first order terms.
718
719 In comparison to the non-limited regime, the calculation of the
720 concentration impacts resulting from different percentage of
721 emission reduction shows non-linear trends (Figure 5). A
722 discontinuity appears at 50% reduction for the abatement of
723 industrial emissions. This discontinuity corresponds to a change of
724 chemical regime. Below the 50% reduction level , the quantity of
725 NH_3 is not sufficient to feed the reactions with NO_2 and SO_2 (with
726 no SO_2 reduction, 50 moles of NO_2 and 50 moles of SO_2 would
727 require 150 moles of NH_3 but only 100 are available) while beyond
728 this 50% reduction level the quantity of NH_3 is then enough to feed
729 the reactions with NO_2 and SO_2 (with 50% SO_2 reduction, 50 moles
730 of NO_2 and 25 moles of SO_2 requires 100 moles of NH_3).
731
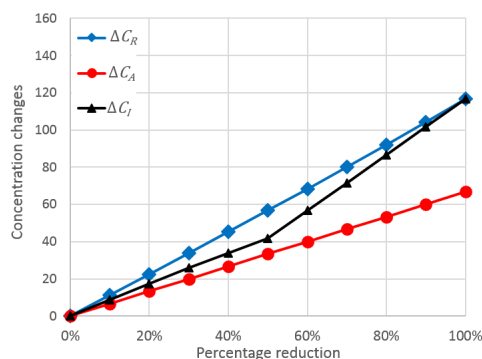
732
733 Figure 5: Evolution of the concentration changes resulting from
734 different percentage of source abatement (Top down approach) for
735 the three sectors (Residential, Agricultural and Industrial).

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25

736
737 The methodologies presented in this section aim at decomposing the
738 impact of an ensemble of sources into different terms attributed to
739 each individual sources. The terms computed by methodologies
740 designed for source apportionment (like TAG) are named source
741 contributions. Their sum is always equal to the combined impact of
742 all sources. On the other hand, the terms computed by sensitivity
743 analysis represent the emission change of each individual source and
744 their sum is equal to the combined impact of all sources only if the
745 relationship between emissions and concentrations is linear (see
746 Section 2). In non-linear situations, the source contributions
747 computed for source apportionment and the source impacts
748 computed for sensitivity analysis are different (see Figure 4 where
749 column 2 shows different results than column 3 or 4). Non-linearity
750 also imply that the calculation of the source impacts depends from
751 the bounds used to estimate the concentration changes (denoted "H"
752 and "L" in Section 4). The BF-BU and BF-TD (columns 3 and 4 in
753 Figure 4) give different results because they are not using the same
754 reference level ("L" for the BU and "H" for the TD as defined in
755 Section 4). Moreover, the results depend from the percentage of
756 emission changes applied to calculate the source impacts as
757 demonstrated by the different source impacts computed with the BF-
758 TD for 100% and 25% emission reductions (columns 4 and 5 in
759 Figure 4). We expect that lower percentage emission reductions
760 generate less non-linearity and lead to a better agreement between
761 the BF-TD and the DDM method (columns 5 and 6 in Figure 4).
762 In synthesis, the second example illustrates that all the
763 methodologies tested to find source contributions and source
764 impacts give different results when the relationship emissions-
765 concentrations is non-linear. It is therefore important to determine
766 the degree of non-linearity in order to understand the range of
767 applicability of each methodology and apply it for the right purpose.

768 **6. Conclusions**

769
770 When pollutant concentrations exceed the thresholds set in the
771 legislation, competent authorities must take actions to abate
772 pollution. In the European legislation (Directive 2008/50/EC) both

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773   source apportionment and planning are requested when designing air
774   quality plans. In practice, source apportionment is often used for
775   planning purposes. It is indeed intuitive to use source apportionment
776   to detect the activity sectors that need to be tackled in priority in an
777   air quality plan. On the other hand, sensitivity analysis is often used
778   as an approach to derive source contributions (e.g. SHERPA,
779   FASST, GAINS…).
780
781   In this work, we compared the two above mentioned approaches and
782   investigated their domain of application. While sensitivity analysis
783   refers to impacts to characterize the concentration change resulting
784   from a given emission change, source apportionment aims to
785   quantify contributions, by attributing a fraction of the pollutant
786   concentration to each emission source. In the case of linear
787   relationships between concentration and emissions, impacts and
788   contributions are equivalent concepts. Source apportionment may
789   then be used for air quality planning purposes and vice versa,
790   sensitivity analysis may be used for quantifying sources
791   contributions.
792
793   In many cases, however, linearity is not a valid assumption. In such
794   cases, sensitivity approaches cannot be used to retrieve source
795   apportionment information, unless non-linear interaction terms are
796   explicitly accounted for. On the other hand, source apportionment
797   approaches (e.g. tagged species approach) intrinsically account for
798   these non-linear interactions into their source contributions. But
799   because it mix interaction terms and impacts, which may react in
800   opposite directions, source apportionment should not be used to
801   evaluate the impact of abatement strategies
802   Even when using sensitivity analysis for planning, it is important to
803   note that, under non-linear conditions, the calculated impacts will
804   only provide information for the exact conditions that are
805   considered. Impacts for an emission reduction of 50% are only valid
806   for exactly that percentage of reduction and extrapolation to air
807   quality planning with any other emission reduction levels would be
808   inappropriate, unless additional scenarios are tested. Along the same
809   line of reasoning, the importance of the non-linear interaction terms
810   (among precursors) should be quantified as well when assessing the

811 impact of more sources or precursors. Finally, these non-linear
812 interaction terms are in most cases not constant with the emission
813 reduction intensities, which would imposes the further need to
814 quantify them for different levels of emission reduction.
815
816 Fortunately, not all cases are so complex to require the full
817 quantification of all non-linear interaction terms. Thunis et al. (2015)
818 showed that for yearly average relationships between emission and
819 concentration changes, linearity is a realistic assumption, implying
820 the possible use of source apportionment and sensitivity analysis for
821 both purposes. Some integrated assessment tools (e.g. GAINS,
822 SHERPA) take advantage of this assumption to retrieve source
823 apportionment information from calculated CTM sensitivities.
824 Although non-linearities are important for short-term time averages
825 (e.g. daily means, episodes), they are likely not associated to every
826 process. For instance, non-linear interactions are expected to be
827 more relevant for secondary pollutants, especially under limited
828 regimes. The challenge consists, therefore, in screening the system
829 for significant non-linearities and account for them by calculating
830 explicitly the relevant non-linear interaction terms.
831
832 One main strength of source apportionment approaches is to provide
833 contribution estimates that can be cross validated with source
834 apportionment derived from measurements (i.e. receptor modelling,
835 for a detailed description see e.g. Belis et al., 2013). This step is
836 crucial for the evaluation of chemistry transport models.
837

838 ## 7. Code availability
839 No specific code is attached to this work as all presented examples
840 can easily be replicated.
841

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843

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