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 decoupled direct method (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 and sensitivity analysis may be used indifferently for both air quality planning purposes and 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. A quantification of the potential non-linearities should therefore be the first step prior to source apportionment or planning applications, to prevent any limitations in their use. When non-linearity is mild, these limitations may however be acceptable in the context of the other uncertainties inherent to complex models.

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36
37 Moreover, when using sensitivity analysis for planning, it is
38 important to note that, under non-linear circumstances, the
39 calculated impacts will only provide information for the exact
40 conditions (e.g. emission reduction share) that are simulated.
41
42
43 **Keywords:** source apportionment, sensitivity analysis, abatement strategies,
44 air quality planning
45

1. Introduction

47
48 When pollutant concentrations exceed the thresholds set in the
49 legislation, competent authorities must take actions to abate
50 pollution. Those abatement strategies consist in reducing the
51 precursor's emission of the different activity sector to reduce
52 pollutant concentrations but they are challenging to design because
53 of the complex relationships that link emissions and pollutants.
54 Indeed, the concentration of a pollutant at a given location generally
55 results from direct emissions and from interactions in the
56 atmosphere among different emission precursors, emitted by a
57 variety of sources. For example, particulate matter (denoted here as
58 PM) results from the interaction and combination of five different
59 precursors (PPM, NO_x, SO_2, NH_3, and VOC) which can be emitted
60 by different activity macro-sectors (e.g. residential, transport,
61 industrial and agriculture [Seinfeld and Pandis, 2016]).
62 Two different approaches are currently used to support air quality
63 decision makers: source apportionment and sensitivity analysis.
64
65 • Source apportionment quantifies the **contribution** of an emission
66 source (or precursor) to the concentration of one pollutant at one
67 given location.
68 • Sensitivity analysis estimates the **impact** on pollutant
69 concentration that results from a change of one or more emission
70 sources.
71

72 In practice, source apportionment is often used for planning
73 purposes. It is indeed intuitive to use source apportionment to detect
74 the activity sectors that need to be tackled in priority in an air quality
75 plan. On the other hand, sensitivity analysis is often used as an
76 approach to derive source contributions,e.g. SHERPA (Thunis et al.
77 2017), FASST (Crippa et al. 2017), GAINS (Kiesewetter et al.
78 2015)…
79 The main objective of this work is to review the existing
80 methodologies, identify key differences and stress their implicit
81 limitations. We will particularly focus on the differences between
82 concentration "impacts" (sensitivity) and "contributions" (source
83 apportionment) obtained with different methodologies. We make use
84 of a simple theoretical example to compare the approaches, highlight
85 differences and potential implications in terms of policy. In the
86 following sections, we analyze first how these methodologies work
87 in a simple linear case before generalizing it to more complex non-
88 linear situations.
89

90 ## 2. Linear simplification and implications

91

92 Let's consider C a pollutant concentration at one location that is a
93 function of three variables (E_1, E_2 and E_3), i.e. the emissions of three
94 precursors or sources within a given domain: $C = C(E_1, E_2, E_3)$. For
95 a linear relationship between the function C and the three variables
96 E_1, E_2 and E_3, we can write:
97

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

98
99 where P_1, P_2 and P_3 are three constant coefficients.
100
101 On the other hand, the sensitivity of the concentration to a change of
102 a given emission source can be quantified via partial derivatives. For
103 expression (1) this gives:
104

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

105

106 In Clappier et al. (2017) the coefficients (P_1, P_2 and P_3) are referred
107 to "potencies". The authors used this concept of "potencies" to
108 analyze the model response to emission changes in different European
109 countries.

110

111 The consequences of a linear relationship between concentration and
112 emission sources are twofold:
113 1) all higher order derivatives (order 2 and beyond) are null, including
114 those involving two or more emission sources (crossed derivatives) as
115 the impact of a change in one emission source is independent from all
116 others.
117 2) the first-order partial derivatives are constant and can therefore be
118 calculated with finite differencing, between any couple of emission
119 levels, for example a base case (denoted BC) and a background
120 (denoted as 0).
121 The potency equations then read as:

122

123
$$P_1 = \frac{\Delta C_0^{BC_1}}{E_1^{BC}} \; ; \; P_2 = \frac{\Delta C_0^{BC_2}}{E_2^{BC}} \; ; P_3 = \frac{\Delta C_0^{BC_3}}{E_3^{BC}}$$

124

125 with $\quad \Delta C_0^{BC_1} = C(E_1^{BC}, 0, 0) - C(0,0,0),$
126 $\quad\quad\quad \Delta C_0^{BC_2} = C(0, E_2^{BC}, 0) - C(0,0,0)$
127 $\quad\quad\quad \Delta C_0^{BC_3} = C(0, 0, E_3^{BC}) - C(0,0,0)$

128

129 Together with "potencies", Clappier et al. (2017) also introduce the
130 concept of "potential", defined as the concentration change resulting
131 from a total reduction of the emissions (from BC to 0). The "potential"
132 can be calculated via relation (1) applied between the base-case and
133 background levels as:

134

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

135

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

137

138 Equation (2) can directly be used for source apportionment purpose,
139 with $\Delta C_0^{BC_1}$ the concentration change resulting from a total reduction
140 of the emission source (or precursor) E_1, reflecting the contribution
141 of E_1 to the base case concentration. Similarly $\Delta C_0^{BC_2}$ and $\Delta C_0^{BC_3}$ are
142 the contributions of E_2 and E_3 . Equation (2) shows that, in the
143 linear case, the concentration change resulting from a simultaneous
144 reduction of all emission sources (ΔC_0^{BC}) is equal to the sum of the
145 emission sources contributions.
146
147 In the next sections, we will explore how this simple conclusion
148 changes when non-linear relationships are considered. In particular,
149 we will assess which implications (and limitations) these non-
150 linearities have in terms of source apportionment and sensitivity
151 analysis.
152

153 **3. Brute Force method**
154
155 The "brute force" method consists in estimating the concentration
156 change by performing and subtracting two simulations, one with and
157 the second without a specific emission source to be analysed
158 (Blanchard, 1999; Yarwood et al., 2004).
159 In non-linear situations, the concentration change resulting from a
160 set of emission sources is not anymore equivalent to the sum of the
161 concentration changes resulting from emission sources changed
162 individually. In the following, we will refer to the work of Stein and
163 Alpert (1993) who proposed an approach to decompose an overall
164 impact into single (one emission source only) and combined
165 (multiple emission sources) impacts.
166
167
168   3.1. Bottom-up formulation
169
170 We consider here three precursor's emission E_1, E_2 and E_3 that are
171 changing from a low (denoted as "L") to a high level (denoted as
172 "H"). In a bottom-up approach, the low emission level is chosen as
173 the reference. With these definitions and notations, the impact on

174 concentration resulting from a change of one only of the three
175 precursor's emissions can be written as follows:
176

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

177
178 While the impact on concentration resulting from the simultaneous
179 changes of two or three precursor's emissions would be written as:
180

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

181 Using a similar notation, the decomposition of Stein and Alpert
182 (1993) applied to 2 variables (E_1 and E_2) would read as:
183

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

184
185

186 where $\Delta C_{\bar{L}}^{H_1}$ and $\Delta C_{\bar{L}}^{H_2}$ are the impacts induced by the change in
187 emission sources E_1 and E_2 taken independently and $\Delta C_{\bar{L}}^{H_1,H_2}$ the
188 impact induced from E_1 and E_2 taken simultaneously.
189
190 It is clear from (3) that the impact of a simultaneous change of two
191 emission sources is not equivalent to the sum of the individual
192 impacts, as highlighted by the additional term $\hat{C}^{int}$. This term that
193 quantifies the interaction between the two emission sources can be
194 calculated using (3) as:
195

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

196
197 The Stein and Alpert formulation can similarly be applied with 3
198 emission sources:

199

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

200

201 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
202 resulting from single emission changes in the sources and

203

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

204

205 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
206 can be further decomposed via equation (4). $\hat{C}_{\bar{L}}^{H_1,H_2,H_3}$ is the triple
207 interaction term (between E_1, E_2, E_3) which can be decomposed by
208 combining (5) and (6) as:

209

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

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

210
211   3.2. Top-down formulation
212
213 In a top-down formulation, the highest emission level is chosen as
214 reference. The Stein Alpert formulation for three precursors can then
215 be expressed similarly to the bottom-up formulation as:
216

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

217

218 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
219 by reducing E_1, E_2 and E_3 independently whereas $\hat{C}^{int}$ is the
220 interaction term which itself can be decomposed into a series of
221 double interactions and a triple interaction terms:

222

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

223

224 It is important to stress that the top-down single impacts are not
225 equivalent to their bottom-up counterparts. The relation between

226 these bottom-up and top-down impacts can be expressed as (here for
227 the case of E$_3$):
228

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

229
230
231

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

232
233
234

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

235
236 Using equations (3), (4), (5) and (6), equation (9) can be re-
237 expressed as:
238

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

239
240 In other words, the top-down impact on concentration of an emission
241 source (obtained by switching off the emission source while all
242 others remain unchanged) is not equivalent to its bottom-up
243 counterpart, (obtained by switching on the emission source while all
244 others are switched off). Relation (10) indeed clearly shows that
245 additional interaction terms need to be considered. The implications
246 resulting from these differences are highlighted in Section 5 in
247 which some theoretical examples are described.
248

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

250
251   4.1. Tagged species techniques
252
253 Equation (2) shows that, when the relationship between concentration
254 and several emission sources is linear, the contribution of a specific
255 source (source apportionment) can be computed as the impact on
256 concentration obtained by a full reduction of this source (sensitivity).
257 Moreover, the sum of the impacts on concentration obtained by
258 reduction of the single sources ($\Delta C_0^{BC_1} + \Delta C_0^{BC_2} + \Delta C_0^{BC_3}$) is equivalent to

the impact on concentration resulting from a simultaneous abatement of all sources (ΔC_0^{BC}). In such a case, the concentration impacts are equal to source contributions and source apportionment and sensitivity analysis lead to similar results. This is not the case, however, when the relationship between concentrations and emissions is nonlinear. In their approach, Stein and Alpert express the difference between the impact caused by a simultaneous abatement and the sum of the impacts caused by individual abatement as interactions among the different sources. The Stein and Alpert formulation applied between the base-case and background levels is very close to equation (2) but with an additional term that accounts for interactions:

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

Because the interaction terms cannot not be attributed to a single emission source as they represent the interaction between two and more emission sources, the Stein and Alpert methodology does not allow identifying the full contribution of each individual source. It cannot therefore be used for source apportionment purpose, unless the interaction terms are negligible as in the linear case.

Unlike the Stein and Alpert methodology, the tagged species methodology is designed for source apportionment purposes. This methodology tags each precursor and quantifies its contribution (in terms of mass) to the pollutant concentration.

Tagged algorithms are implemented in several chemical transport model systems (Yarwood et al., 2004; Wagstrom et al., 2008, ENVIRON, 2014; Bhave et al., 2007; Wang et al., 2009; Kranenburg et al., 2013).

In tagging approaches, the effect of the full reduction of all sources is directly expressed as the sum of the source contributions:

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

where δC_1, δC_2, δC_3 are the contributions of sources E_1, E_2 and E_3 resulting from the tagged species approach resolution.

297 Tagging methodologies split the interaction terms into fractions and
298 attribute these fractions to the source contributions, on the basis of
299 mass weighting factors.
300
301 $$\delta C_1 = \Delta C_0^{BC_1} + \alpha \hat{C}^{int}$$
302
303
304 Because the tagged species approach mixes interaction terms and
305 single concentration impacts into sources contributions, it is not
306 suitable to estimate the effect of emission reduction when non-
307 linearities are present (Burr and Zhang 2011a, 2011b). Indeed these
308 two types of terms may react in very different ways to emission
309 reductions. This fact is detailed in the examples provided below.
310 On the other hand, the strength of this method is that it allows for a
311 direct comparison of the source contributions with measurements (or
312 measurement based methods like receptor models).
313 Note that similar tagging methods are also used in the frame of
314 climate-chemical studies at global scale (e.g. Horowitz and Jacob,
315 1999; Lelieveld and Dentener, 2000; Meijer et al., 2000; Grewe, 2004;
316 Gromov et al., 2010; Butler et al.,2011; Emmons et al., 2012; Grewe
317 et al., 2012; Grewe et al., 2017).
318
319   4.2. DDM
320
321 The decoupled direct method (DDM) is designed to calculate
322 directly sensitivities to emission changes (Dunker et al., 1984;
323 Dunker et al., 2002). It aims to compute the first order derivatives
324 (which correspond to the potencies mentioned in section 2).
325
326 $$\frac{\partial C}{\partial E_1} \;;\; \frac{\partial C}{\partial E_2} \;;\; \frac{\partial C}{\partial E_3}$$
327
328 The Taylor formula is applied at first order to calculate the
329 concentration change between two emission levels (denoted H and
330 L).
331

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

with $\quad \Delta E_1 = E_1^H - E_1^L, \Delta E_2 = E_2^H - E_2^L, \Delta E_3 = E_3^H - E_3^L$

In the linear case, the first order derivatives are constant and the first order approximation of the Taylor formula gives the exact expression of the impact on concentration of an emission change between H and L. When the emission-concentration relationship is nonlinear, the first derivatives are not constants. The first order Taylor formula cannot take into account the nonlinear effects. It is a linear approximation based on derivatives computed at a given emission reference level (level H in our example). The estimation of the impact on concentration of an emission change between H and L is accurate enough if level L is close enough to level H.

HDDM is another method (Hakami et al., 2003) which aims to increase the accuracy of the DDM method by computing second order derivatives.
DDM (and HDDM) gives similar information to the Stein Alpert formulation applied with the Brute force top-down approach (because the reference level is H). For the same reason than for the Stein Alpert approach, these two methods are suitable for source apportionment purpose only if the relation between concentration and emission is close to linearity.

DDM (and HDDM) approximates the impact on concentration from an emission change between the two level H and L, using derivatives computed at level H. This impact is accurate enough if the level L is close enough to the reference level H.
Dunker (2015) showed how to use first-order sensitivity to determine source contributions between two model cases, e.g., to apportion the difference between the current atmosphere (and natural conditions) to specific human activities. Along the same lines, Simon et al. (2013) used first order sensitivity to construct emission response surfaces. To cope with potential non-linearities and the need to compute higher-order derivatives, a powerful

368 alternative is to compute first-order sensitivities at several emission
369 levels.
370
371
372 **5. Example**
373
374 In this section, examples are designed to illustrate the differences in
375 terms of contribution and impact estimates when the approaches
376 discussed previously are used. In these examples, we focus on the
377 formation of particulate matter (PM) in the atmosphere and only
378 consider three formation processes: direct emissions (primary PM
379 denoted as PPM); formation through reactions with nitrogen oxides
380 (NO_2) and ammonia (NH_3) and formation through reactions with
381 sulphur oxide (SO_2) and NH_3.
382
383 $PPM \longrightarrow PM$

384 $2NO_2 + H_2O + \frac{1}{2}O_2 \longrightarrow 2HNO_3$

385 $2NH_3 + 2HNO_3 \longrightarrow 2NH_4NO_3$

386 $SO_2 + H_2O + \frac{1}{2}O_2 \longrightarrow H_2SO_4$

387 $2\,NH_3 + H_2SO_4 \longrightarrow (NH_4)_2SO_4$

388
389 These reactions pathways are summarised by the following system
390 of reactions:
391
392 $PPM \rightarrow PM[PPM]$

393 $NO_2 + NH_3 \rightarrow PM[NH_4NO_3]$

394 $SO_2 + 2\,NH_3 \rightarrow PM[(NH_4)_2SO_4]$

395
396 This system is further simplified by assuming that all reactions have
397 comparable kinetics (reaction speed) and have reached their
398 equilibrium. From these three reactions, 1 PM mole can be produced
399 by 1 PPM mole, by the combination of 1 NH_3 and 1 NO_2 moles or
400 by the combination of 1 SO_2 and 2 NH_3 moles.

401

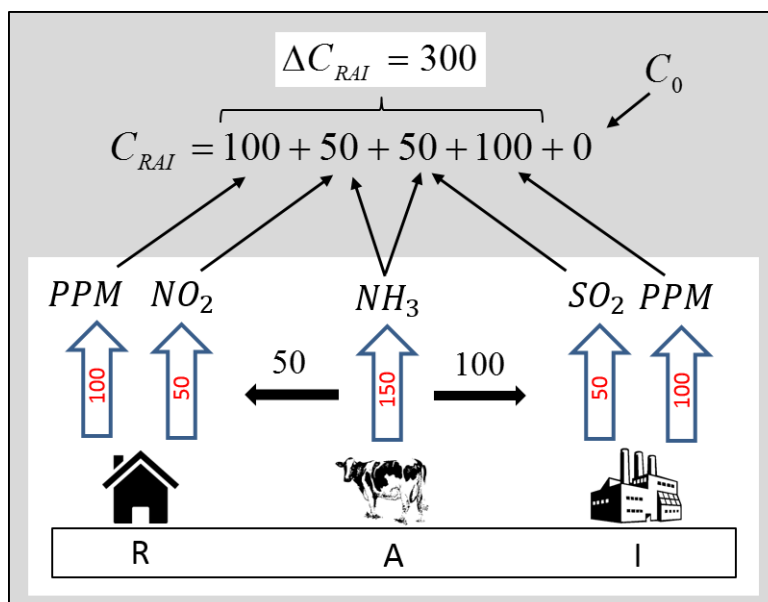
402 We also limit our examples to emissions from three activity sectors.
403 The residential sector (R) only emits PPM and NO_2, the agricultural
404 sector (A) only emits NH_3 and the industrial sector (I) only emits
405 PPM and SO_2 (Figure 1). We assume for convenience that no
406 background pollution is present (i.e. there is no PM when all
407 emissions are zero). Two situations are considered: a "non-limited
408 regime" where the NH_3 quantity is sufficient to react with all moles
409 of NO_2 and SO_2 and a "limited regime" where the NH_3 quantity of is
410 not sufficient to react with all moles of NO_2 and SO_2.

411

412   **5.1. Non-limited regime**

413

414 In this first example, the quantity of precursors (in terms of mass) is
415 large enough to feed all reactions. The agricultural sector emits 150
416 NH_3 moles which can react with 50 NO_2 moles emitted by the
417 residential sector and 50 SO_2 moles emitted by industrial sector. 100
418 PPM moles are emitted by the residential sector as well by the
419 industrial sector (Figure 1).

420

421

422 Figure 1: Example of PPM, NO_2, SO_2 and NH_3 emissions released
423 by three activity sectors: residential (R), agricultural (A) and
424 industrial (I). For convenience, no units are associated to emissions
425 and concentrations
426
427 Let's first calculate the PM concentration produced with and without
428 each of the sources:
429
430 No source: C_0 is the PM concentration obtained when all emissions
431 are set to zero. Since we assumed a zero background pollution, $C_0 =$
432 0.
433
434 One source only: C_R (resp. C_A and C_I) is the PM concentration
435 reached when only the residential (resp. agricultural and industrial)
436 sector releases emissions:
437   - $C_R = 100$ produced by PPM emissions (NO_2 emissions do not
438   produce PM as no NH_3 is available).
439   - $C_A = 0$ because NO_2 and SO_2 are not available to react with NH_3.
440   - $C_I = 100$ produced by the PPM emissions (SO_2 emissions do not
441   produce PM as no NH_3 is available).
442
443 Two sources: C_{RA} C_{RI}, and C_{AI} are the concentrations obtained when
444 two (out of three) activity sectors release their emissions
445 simultaneously (the RA subscripts correspond to residential and
446 agriculture, RI to residential and industrial, AI to agriculture and
447 industrial):
448   - $C_{RA} = 150$: 100 produced by PPM emissions from the residential
449   sector and 50 produced by the 50 NO_2 released by the residential
450   sector reacting with the 50 NH_3 emitted by agriculture (100 NH_3
451   moles remain unused).
452   - $C_{RI} = 200$: 100 produced by PPM emissions from the residential
453   sector and 100 produced by PPM emissions from the industrial
454   sector.
455   - $C_{AI} = 150$: 100 produced by PPM industrial emissions and 50
456   from the combination of 50 SO_2 (industry) and 100 NH_3
457   (agriculture).
458

459   Three sources: C_{RAI} is the concentrations obtained when all
460   emissions are released simultaneously.
461   - $C_{RAI} = 300 : 200$ from PPM (residential and industry), 50 from
462   reaction between NO_2 and NH_3 and 50 from reaction between SO_2
463   and NH_3.
464
465   **Brute-force Bottom-up (BF-BU) method**
466
467   The contribution of each activity sector is calculated as the
468   concentration change resulting from a 100% emission increase from
469   the lowest emission level (previously denoted "L" or background) to
470   the highest level (denoted as "H", the base case C_{RAI} obtained with
471   all emissions)
472
473   In a bottom-up approach, the concentration associated with the
474   lowest emission level is considered as the reference. Concentration
475   impacts are then computed by the difference between any situation
476   (e.g. one, two or three sources present) and this reference.
477

478   - with one source: $\Delta C_R^{BU} = C_R - C_0 = 100$
479 $\Delta C_A^{BU} = C_A - C_0 = 0$
480 $\Delta C_I^{BU} = C_I - C_0 = 100$
481

482   - with two sources: $\Delta C_{RA}^{BU} = C_{RA} - C_0 = 150$
483 $\Delta C_{RI}^{BU} = C_{RI} - C_0 = 200$
484 $\Delta C_{AI}^{BU} = C_{AI} - C_0 = 150$
485

486   - with three sources: $\Delta C_{RAI} = C_{RAI} - C_0 = 300$
487

488   To calculate the interaction terms, we use the Stein-Alpert
489   formulation using Eq. (5) and Eq. (6):
490

491   $\Delta C_{RAI} = \Delta C_R^{BU} + \Delta C_A^{BU} + \Delta C_I^{BU} + \hat{C}_{RA}^{BU} + \hat{C}_{RI}^{BU} + \hat{C}_{AI}^{BU} + \hat{C}_{RAI}^{BU}$
492

493   from which the interaction terms are obtained by application of (4)
494   and (6):
495

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

498 $\quad \hat{C}_{AI}^{BU} = \Delta C_{AI}^{BU} - \Delta C_{A}^{BU} - \Delta C_{I}^{BU} = 50$

499 $\quad \hat{C}_{RAI}^{BU} = \Delta C_{RAI} - \Delta C_{R}^{BU} - \Delta C_{A}^{BU} - \Delta C_{I}^{BU} - \hat{C}_{RA}^{BU} - \hat{C}_{RI}^{BU} - \hat{C}_{AI}^{BU} = 0$

500

501 As can be seen from this example, the system behaves non-linearly

502 and the interaction terms (e.g. $\hat{C}_{RA}^{BU}$) are not-zero. Moreover the sum

503 of the individual impacts ($\Delta C_{R}^{BU} + \Delta C_{A}^{BU} + \Delta C_{I}^{BU} = 200$)

504 underestimates the overall impact ($\Delta C_{RAI} = 300$). These results are

505 graphically represented in Figure 2 (third column).

506

507

508 **Brute-force Top-down (BF-TD) method**

509

510 In a BF-TD approach, the higher emission level (base case, C_{RAI}) is

511 the reference and the impact of each activity sector is calculated as

512 the concentration change resulting from a 100% emission decrease

513 (of one, two or three sources) from this reference to the background

514 level.

515

516   - with one source: When all emissions from one sector are reduced

517   (e.g. residential), the other two sector remain active (agricultural

518   and industry). In this case, the Top-down impact is the difference

519   between the base case concentration and the concentration

520   resulting from the agricultural and industrial emissions only. A

521   similar reasoning can be made for all sectors:

522

523 $\quad\quad\quad\quad \Delta C_{R}^{TD} = C_{RAI} - C_{AI} = 150$

524 $\quad\quad\quad\quad \Delta C_{A}^{TD} = C_{RAI} - C_{RI} = 100$

525 $\quad\quad\quad\quad \Delta C_{I}^{TD} = C_{RAI} - C_{RA} = 150$

526

527   - with two sources: The Top-down impact due to a full reduction

528   of two sectors (e.g. residential and agriculture) is similarly

529   computed as the difference between the base case concentration

530   and the concentration resulting from the remaining sector

531   (industry).

532

533 $\quad\quad\quad\quad \Delta C_{RA}^{TD} = C_{RAI} - C_{I} = 200$

534 $\quad\quad\quad\quad \Delta C_{RI}^{TD} = C_{RAI} - C_{A} = 300$

535 $\quad\quad\quad\quad \Delta C_{AI}^{TD} = C_{RAI} - C_{R} = 200$

536
537
538 - with three sources: The impact resulting from the simultaneous
539 reduction of all three sources is similar in the Top-down and
540 Bottom-up approaches:
541
542
$$\Delta C_{RAI} = C_{RAI} - C_0 = 300$$

543
544 The interaction terms can be obtained in a similar way to the
545 bottom-up approach by using the Stein and Alpert formulation for
546 ΔC_{RAI}:
547
548
$$\Delta C_{RAI} = \Delta C_R^{TD} + \Delta C_A^{TD} + \Delta C_I^{TD} + \hat{C}_{RA}^{TD} + \hat{C}_{RI}^{TD} + \hat{C}_{AI}^{TD} + \hat{C}_{RAI}^{TD}$$

549
550 The interaction terms are given by:
551
552
$$\hat{C}_{RA}^{TD} = \Delta C_{RA}^{TD} - \Delta C_R^{TD} - \Delta C_A^{TD} = -50$$
553
$$\hat{C}_{RI}^{TD} = \Delta C_{RI}^{TD} - \Delta C_R^{TD} - \Delta C_I^{TD} = 0$$
554
$$\hat{C}_{AI}^{TD} = \Delta C_{AI}^{TD} - \Delta C_A^{TD} - \Delta C_I^{TD} = -50$$
555
$$\hat{C}_{RAI}^{TD} = \Delta C_{RAI} - \Delta C_R^{TD} - \Delta C_A^{TD} - \Delta C_I^{TD} - \hat{C}_{RA}^{TD} - \hat{C}_{RI}^{TD} - \hat{C}_{AI}^{TD} = 0$$

556
557 With this approach, a non-linear behavior is also observed and
558 interaction terms are not zero. It is also interesting to note that the
559 triple interaction term ($\hat{C}_{RAI}^{TD}$) is null. The sum of the individual
560 impacts ($\Delta C_R^{TD} + \Delta C_A^{TD} + \Delta C_I^{TD} = 400$) overestimates the overall
561 impact ($\Delta C_{RAI} = 300$).We further discuss these aspects at the end of
562 this section. These results are graphically represented in Figure 2
563 (fourth and fifth columns).
564
565
566 **Tagged species approach**
567
568 Compared to Brute-force, the tagged species approach calculates the
569 share of each source to the overall concentration change. These
570 shares are referred to as contributions and have the main property
571 that the sum of the individual contributions is equal to the overall
572 concentration impact, by definition, i.e.:
573
574 $\Delta C_{RAI} = \delta C_R^{TAG} + \delta C_A^{TAG} + \delta C_I^{TAG}$

575	
576	
577	The sector contributions are computed by tracking the mass of their
578	emitted species contributing to PM formation (in our example:
579	$PM[PPM]$, $PM[NH_4NO_3]$ and $PM[(NH_4)_2SO_4]$)
580	
581	- $PM[PPM]$ is formed from PPM. The 100 moles from the residential
582	sector lead to 100 moles of PM. The same applies to the 100 moles
583	from industry.
584	
585	- $PM[NH_4NO_3]$ is formed by combination of NH_3 and NO_2. The
586	share between these two contributions is obtained by application
587	of stoichiometric molar mass ratios:
588	
589	$$a_1 = \frac{[NO_3]_m}{[NO_3]_m + [NH_4]_m} = 0.78$$
590	
591	In our example, 50 moles of $PM[NH_4NO_3]$ are formed by
592	combination of NO_2 (50 moles) from the residential sector and
593	NH_3 (50 moles) from agriculture. The contribution attributed to
594	NO_2 is $50 \times a_1$ whereas the contribution attributed to NH_3 is
595	$50 \times (1 - a_1)$.
596	
597	
598	- $PM[(NH_4)_2SO_4]$ is formed by combination of NH_3 and SO_2. The
599	following stoichiometric mass ratio is used:
600	
601	$$a_2 = \frac{[SO_4]_m}{[SO_4]_m + 2[NH_4]_m} = 0.73$$
602	
603	The contribution attributed to SO_2 is $50 \times a_2$ whereas the
604	contribution attributed to NH_3 is $50 \times (1 - a_2)$.
605	
606	The contribution of each sector is then obtained as the sum of their
607	precursor contribution shares as follows:
608	
609	$\delta C_R^{TAG} = 100 + 50 \times a_1 = 138.7$
610	$\delta C_A^{TAG} = 50 \times (1 - a_1) + 50 \times (1 - a_2) = 24.9$

611 $\delta C_I^{TAG} = 100 + 50 \times a_2 = 136.4$

612

613 By definition the sum of the contributions ($\delta C_R^{TAG} + \delta C_A^{TAG} +$

614 $\delta C_I^{TAG} = 300$) is exactly equal to the overall concentration impact

615 ($\Delta C_{RAI} = 300$).

616

617 Note that a decomposition of the non-linear interaction terms

618 obtained in the top-down or bottom-up approach, using the above

619 stoichiometric ratios would lead to similar results than the tagged

620 approach. These results are graphically represented in Figure 2

621 (second column).

622

623

624 **DDM**

625

626 In this methodology, delta concentrations and interaction terms are

627 estimated with first order partial derivatives computed from the

628 highest emission level (base case in our example). Being a

629 sensitivity approach using level H as reference, DDM shows clear

630 analogies with the BF-TD.

631

632 $$\left.\frac{\partial C}{\partial \alpha_R}\right|_{TD} = 150 \qquad \left.\frac{\partial C}{\partial \alpha_A}\right|_{TD} = 100 \qquad \left.\frac{\partial C}{\partial \alpha_I}\right|_{TD} = 150$$

633

634

635 where α_R, α_A and α_I are percentage emission changes from the base

636 case for the residential, agricultural and industrial sectors.

637 The first-order derivatives are evaluated using finite differencing

638 between the base case and a level characterised by emissions that are

639 10% lower for each activity sector.

640 The concentration changes resulting from a 100% emission

641 reduction (i.e. between the base case and the zero emission case)

642 could be estimated by setting α_R, α_A and α_I to unity.

643

644 $$\Delta C_R^{HDDM} = \left.\frac{\partial C}{\partial \alpha_R}\right|_{TD} = 150$$

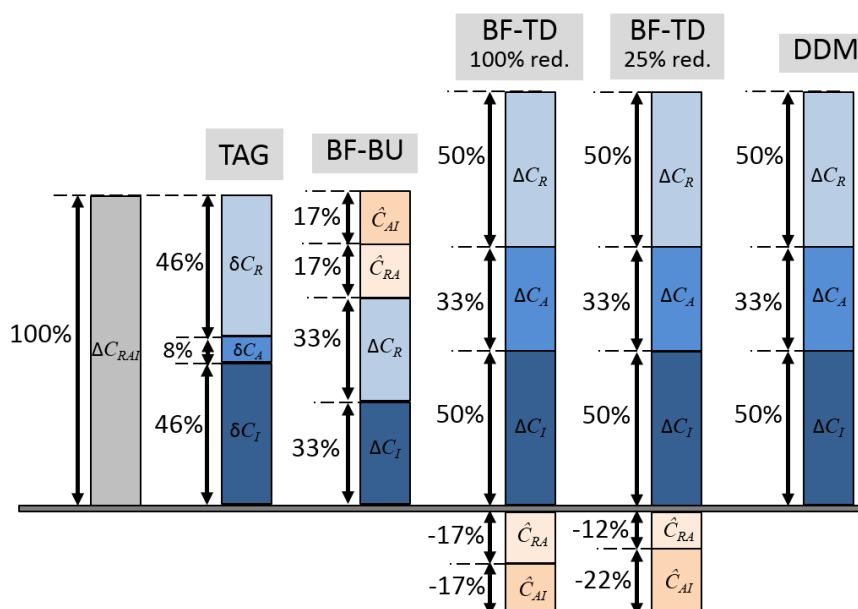
645 $$\Delta C_A^{HDDM} = \left.\frac{\partial C}{\partial \alpha_A}\right|_{TD} = 100$$

646 $\Delta C_I^{HDDM} = \frac{\partial C}{\partial \alpha_I}\bigg|_{TD} = 150$

647

648

649 We see from this last example that both the total PM and the
650 contribution of the sources are then comparable with those obtained
651 with the BF-TD method. Their interpretation is similar (Figure 2,
652 sixth column). In their work, Koo et al. (2009) present a detailed
653 comparison between a DDM and a tagged species approach in a 3-D
654 PM model and show which sensitivities are similar to
655 apportionment, and which are not.

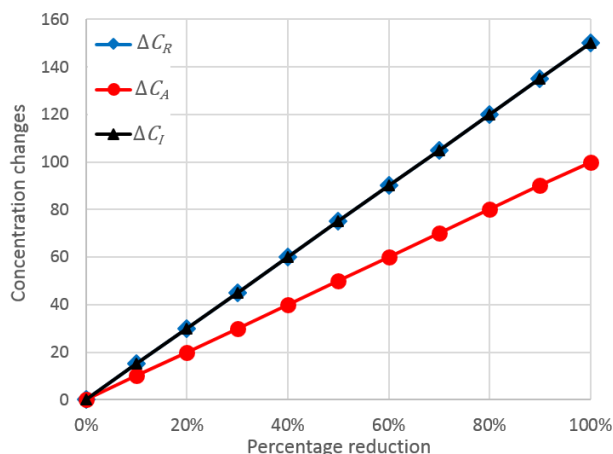
656

657
658 Figure 2: Schematic representation of the allocation of PM to its
659 sources in the non-limited example. The expected total PM is
660 displayed in the grey bar on the left.

661

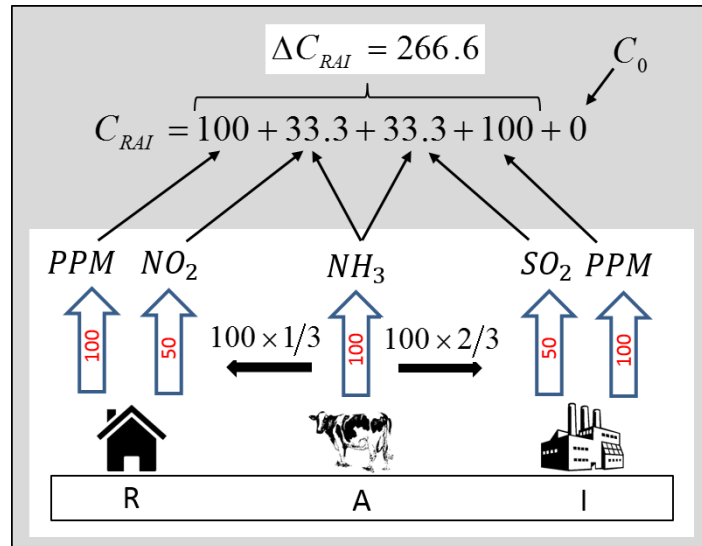
662 **Comparative overview**

663

664 In the linear case (second paragraph) we have seen that a single
665 source contribution can be computed as the impact resulting from a
666 full reduction of this source. However, source contributions and

667   concentration impacts should not be confused as they are different in
668   most situations. The example presented in this paragraph illustrates
669   this clearly for a non-linear system. Indeed the contributions of the
670   single sources computed by the tagged species approach ($\delta C_R^{TAG} =$
671   138 ; $\delta C_A^{TAG} = 24$; $\delta C_I^{TAG} = 136$) differ from the concentration
672   impacts resulting from a total abatement of these single sources
673   computed by the BF-TD ($\Delta C_R^{TD} = 150$; $\Delta C_A^{TD} = 100$; $\Delta C_I^{TD} = 150$)
674   method. Moreover, the sum of the concentration impacts obtained
675   with either the BF-TD approach for single sources does not equal the
676   total concentration impact ($\Delta C_{RAI} = 300$). This is also valid for any
677   selection of two sectors ($\Delta C_R^{TD} + \Delta C_A^{TD} = 250 \neq \Delta C_{RA}^{TD} = 200$). Note
678   that similarly to BF-TD, the concentration impacts computed as
679   increases from the background (BF-BU) show the same behaviour
680   ($\Delta C_R^{BU} + \Delta C_A^{BU} = 100 \neq \Delta C_{RA}^{BU} = 150$).
681
682   Figure 3 shows that the impact on concentration is proportional to
683   the emission reduction indicating that the relationship between
684   emission and concentration changes is linear. However, this example
685   also illustrates the fact that linearity encompasses two aspects: 1)
686   the interaction terms are zero ($\hat{C}^{int} = 0$); 2) the ratios between
687   concentration change and emission changes ($\Delta C / \Delta E$) remain
688   constant, regardless of the calculation bounds (denoted "H" and "L"
689   in Section 4). In the current example the ratios $\Delta C / \Delta E$ are constant
690   (linear trend of ΔC on Figure 3) but the relationship between
691   concentration and emission is not linear because of the non-zero
692   interaction terms (not shown) ($\hat{C}_{RA}^{TD} = -50$ and $\hat{C}_{AI}^{TD} = -50$).
693   However, even with zero interaction terms, we can still observe a
694   non-linear behaviour with the emission reduction percentage. The
695   evaluation of linearity therefore requires two tests: one to quantify
696   the interaction terms and the second to assess the deviation from a
697   linear trend with respect to the emission reduction percentage.
698

699
700 Figure 3: Evolution of the concentration changes resulting from
701 different percentage of source abatement (Top down approach) for
702 the three sectors (Residential, Agricultural and Industrial).
703
704
705

5.2. Limited regime

707
708 This example is similar to the previous, except that the emissions of
709 NH_3 are reduced from 150 to 100 moles.
710 When all sources release emissions, the 100 moles of NH_3 are
711 shared into 100/3=33.3 moles which react with NO_2 to form 33.3
712 moles of $PM[NH_4NO_3]$ and 100×2/3=66.6 moles which react with
713 SO_2 to give 33.3 moles of $PM[(NH_4)_2SO_4]$. Because the mass of
714 NH_3 is not enough to react with all the NO_2 and SO_2 mass, 16.7
715 moles of NO_2 and 16.7 moles of SO_2 remain unused (Figure 4).
716 Note that when the agricultural source is active with one only of the
717 two other sources (residential or industrial) the NH_3 100 moles are
718 then sufficient to consume all the NO_2 or SO_2 and lead to 50 moles
719 of PM in either case.
720

721
722   Figure 4: Example with three sources in an ammonia-limited regime.
723   The mass emitted by each source is expressed in moles.
724
725   The PM concentrations obtained when one or two sources are active
726   are similar to the previous example:
727
728        $C_0 = 0$; $C_R = 100$; $C_A = 0$; $C_I = 100$
729        $C_{RA} = 150$; $C_{RI} = 200$; $C_{AI} = 150$
730
731   But the result differ when all sources are active: $C_{RAI} = 266.6$ (200
732   from PPM (residential industry), 33.3 from reaction between NO_2
733   and NH_3 and 33.3 from reaction between SO_2 and NH_3).
734
735   **Bottom-up brute-force method (BF-BU)**
736
737   The BF-BU approach computes all concentration impacts from the
738   background concentration (C_0). The Stein-Alpert terms are similar to
739   the non-limited case, excepted for ΔC_{RAI} and $\hat{C}_{RAI}$:
740
741        $\Delta C_R^{BU} = 100$ $\hat{C}_{RA}^{BU} = 50$
742        $\Delta C_A^{BU} = 0$ $\hat{C}_{RI}^{BU} = 0$
743        $\Delta C_I^{BU} = 100$ $\hat{C}_{AI}^{BU} = 50$
744        $\Delta C_{RAI} = 266.6$ $\hat{C}_{RAI} = -33.3$

745
746 The limiting effect of NH$_3$ appears only in the negative triple
747 interaction term ($\hat{C}_{RAI}$). These results are graphically represented in
748 Figure 5 (third column).

749
750 **Top-down brute-force method (BF-TD)**

751
752 The Top-down approach uses the base case (C_{RAI}) concentration as
753 reference to compute the concentration impact. In this case, all
754 Stein-Alpert terms are different from the non-limited regime:

755
756 $\Delta C_R^{TD} = 116.6$ $\hat{C}_{RA}^{TD} = -16.6$
757 $\Delta C_A^{TD} = 66.6$ $\hat{C}_{RI}^{TD} = 33.3$
758 $\Delta C_I^{TD} = 116.6$ $\hat{C}_{AI}^{TD} = -16.6$
759 $\Delta C_{RAI} = 266.6$ $\hat{C}_{RAI} = -33.3$

760
761 These results are graphically represented in Figure 5 (fourth and
762 fifth columns).

763
764 **Tagged approach**

765
766 The contribution of each source is computed similarly to the non-
767 limited regime. The production of 33.3 moles of PM[NH_4NO_3] and
768 33.3 moles of PM[$(NH_4)_2SO_4$] are split among the different sectors
769 using the stoichiometric coefficients a_1 and a_2:

770
771 $\delta C_R^{TAG} = 100 + 33.3 * a_1 = 125.8$
772 $\delta C_A^{TAG} = 33.3 * (1 - a_1) + 33.3 * (1 - a_2) = 16.6$
773 $\delta C_I^{TAG} = 100 + 33.3 * a_2 = 124.2$

774
775 These results are graphically represented in Figure 5 (second
776 column).

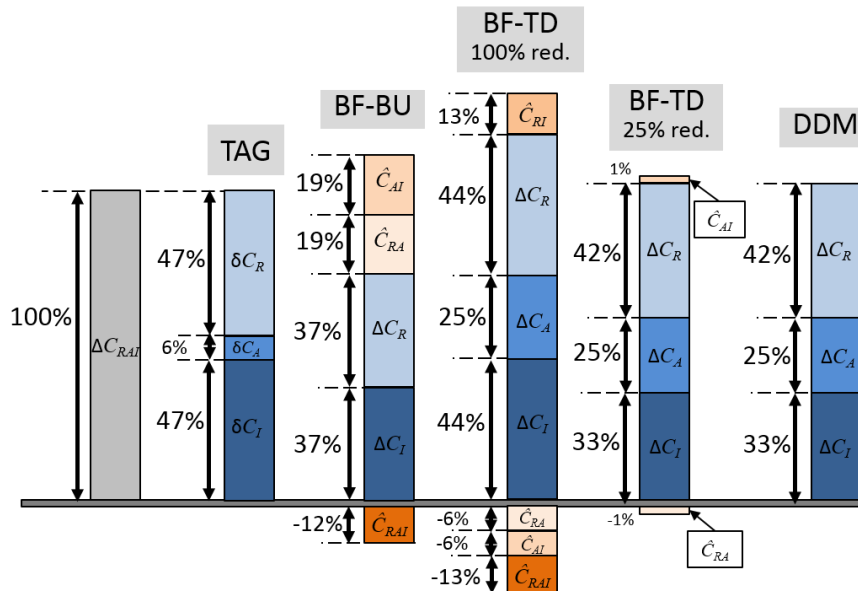
777
778
779 **DDM**
780

781 As shown below, DDM only considers first derivatives, which are
782 not suitable to estimate higher order interaction terms. The
783 calculation of the first order derivatives in this example gives:
784

785 $$\Delta C_R^{HDDM} = \left.\frac{\partial C}{\partial \alpha_R}\right|_{TD} = 111.5$$

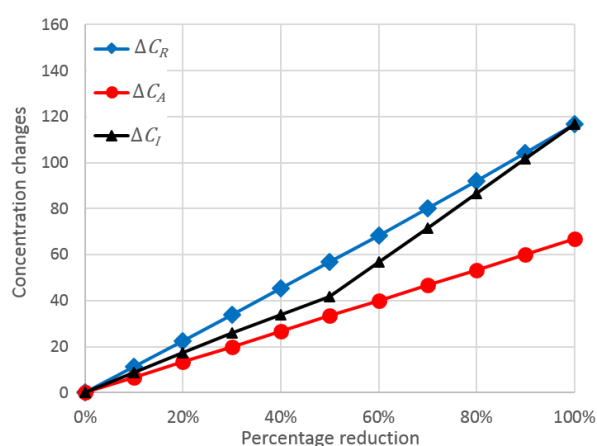
786 $$\Delta C_A^{HDDM} = \left.\frac{\partial C}{\partial \alpha_A}\right|_{TD} = 66.7$$

787 $$\Delta C_R^{HDDM} = \left.\frac{\partial C}{\partial \alpha_I}\right|_{TD} = 88.1$$

788
789 These results are graphically represented in Figure 5 (sixth column).

790
791 Figure 5: Schematic representation of the allocation of PM to its
792 sources in the ammonia-limited example. The expected total PM is
793 displayed in the grey bar on the left.

794
795
796 **Comparative overview**

797
798 The main difference with respect to the non-limited regime is the
799 appearance of a triple interaction term that will also lead to

800 differences between the BF-TD and the DDM approaches, given the
801 fact that the latter only accounts for first order terms.
802 In comparison to the non-limited regime, the calculation of the
803 concentration impacts resulting from different percentage of
804 emission reduction shows non-linear trends (Figure 6). A
805 discontinuity appears at 50% reduction for the abatement of
806 industrial emissions. This discontinuity corresponds to a change of
807 chemical regime. Below the 50% reduction level , the quantity of
808 NH_3 is not sufficient to feed the reactions with NO_2 and SO_2 (with
809 no SO_2 reduction, 50 moles of NO_2 and 50 moles of SO_2 would
810 require 150 moles of NH_3 but only 100 are available) while beyond
811 this 50% reduction level the quantity of NH_3 is then enough to feed
812 the reactions with NO_2 and SO_2 (with 50% SO_2 reduction, 50 moles
813 of NO_2 and 25 moles of SO_2 requires 100 moles of NH_3).
814
815

816
817 Figure 6: Evolution of the concentration changes resulting from
818 different percentage of source abatement (Top down approach) for
819 the three sectors (Residential, Agricultural and Industrial).
820
821
822 The methodologies presented in this section aim at decomposing the
823 impact of an ensemble of sources into different terms attributed to
824 each individual sources. The terms computed by methodologies
825 designed for source apportionment (like TAG) are named source

826   contributions. Their sum is always equal to the combined impact of
827   all sources. On the other hand, the terms computed by sensitivity
828   analysis represent the emission change of each individual source and
829   their sum is equal to the combined impact of all sources only if the
830   relationship between emissions and concentrations is linear (see
831   Section 2). Grewe at al. (2010) and Grewe (2013) who used simple
832   differential equations to reproduce the ozone tropospheric chemistry,
833   also highlighted this point in their work. In non-linear situations, the
834   source contributions computed for source apportionment and the
835   source impacts computed for sensitivity analysis are different (see
836   Figure 5 where column 2 shows different results than column 3 or
837   4). Non-linearity also imply that the calculation of the source
838   impacts depends from the bounds used to estimate the concentration
839   changes (denoted "H" and "L" in Section 4). The BF-BU and BF-
840   TD (columns 3 and 4 in Figure 5) give different results because they
841   are not using the same reference level ("L" for the BU and "H" for
842   the TD as defined in Section 4). Moreover, the results depend from
843   the percentage of emission changes applied to calculate the source
844   impacts as demonstrated by the different source impacts computed
845   with the BF-TD for 100% and 25% emission reductions (columns 4
846   and 5 in Figure 5). We expect that lower percentage emission
847   reductions generate less non-linearity and lead to a better agreement
848   between the BF-TD and the DDM method (columns 5 and 6 in
849   Figure 5).
850   In synthesis, the second example illustrates that all the
851   methodologies tested to find source contributions and source
852   impacts give different results when the relationship emissions-
853   concentrations is non-linear. A quantification of the potential non-
854   linearities should therefore be the first step prior to source
855   apportionment or planning applications, to prevent any limitations in
856   their use. When non-linearity is mild, these limitations may however
857   be acceptable in the context of the other uncertainties inherent to
858   complex models.
859

860   **6. Conclusions**

861

862 In this work, we compared source apportionment and sensitivity
863 approaches and investigated their domain of application. While
864 sensitivity analysis refers to impacts to characterize the
865 concentration change resulting from a given emission change, source
866 apportionment aims to quantify contributions, by attributing a
867 fraction of the pollutant concentration to each emission source. In
868 the case of linear (or close to linear) relationships between
869 concentration and emissions, impacts and contributions are
870 equivalent (or close to) concepts. Source apportionment may then be
871 used for air quality planning purposes and vice versa, sensitivity
872 analysis may be used for quantifying sources contributions.
873
874 In many cases, however, linearity is not a valid assumption. In such
875 cases, sensitivity approaches cannot be used to retrieve source
876 apportionment information, unless non-linear interaction terms are
877 explicitly accounted for. On the other hand, source apportionment
878 approaches (e.g. tagged species approach) intrinsically account for
879 these non-linear interactions into their source contributions. But
880 because it mix interaction terms and impacts, which may react in
881 opposite directions, source apportionment should not be used to
882 evaluate the impact of abatement strategies
883 Even when using sensitivity analysis for planning, it is important to
884 note that, under non-linear conditions, the calculated impacts will
885 only provide information for the exact conditions that are
886 considered. Impacts for an emission reduction of 50% are only valid
887 for exactly that percentage of reduction and extrapolation to air
888 quality planning with any other emission reduction levels would be
889 inappropriate, unless additional scenarios are tested. Along the same
890 line of reasoning, the importance of the non-linear interaction terms
891 (among precursors) should be quantified as well when assessing the
892 impact of more sources or precursors. Finally, these non-linear
893 interaction terms are in most cases not constant with the emission
894 reduction intensities, which would imposes the further need to
895 quantify them for different levels of emission reduction. Calculating
896 sensitivities and interactions at various level of emission reductions
897 seems the only alternative when non-linearities are important. In this
898 respect, new approaches like the path-integral methodology

899 proposed by Grewe et al. (2012) might represent a powerful
900 approach.
901
902 Fortunately, not all cases are so complex to require the full
903 quantification of all non-linear interaction terms. Thunis et al. (2015)
904 showed that for yearly average relationships between emission and
905 concentration changes, linearity is a realistic assumption, implying
906 the possible use of source apportionment and sensitivity analysis for
907 both purposes. Some integrated assessment tools (e.g. GAINS,
908 SHERPA) take advantage of this assumption to retrieve source
909 apportionment information from calculated CTM sensitivities.
910 Although non-linearities are important for short-term time averages
911 (e.g. daily means, episodes), they are likely not associated to every
912 process. For instance, non-linear interactions are expected to be
913 more relevant for secondary pollutants, especially under limited
914 regimes. The challenge consists, therefore, in screening the system
915 for significant non-linearities and account for them by calculating
916 explicitly the relevant non-linear interaction terms.
917
918 One main strength of source apportionment approaches is to provide
919 contribution estimates that can be cross validated with source
920 apportionment derived from measurements (i.e. receptor modelling,
921 for a detailed description see e.g. Belis et al., 2013). This step is
922 crucial for the evaluation of chemistry transport models.
923

924 ## 7. Code availability
925 No specific code is attached to this work as all presented examples
926 can easily be replicated.
927
928
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930

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