

# Source apportionment to support air quality management practices

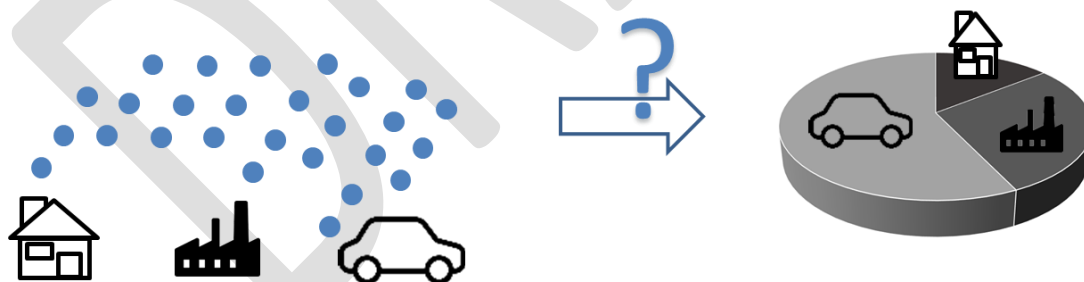
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## A fitness-for-purposes guide

Authors: P. Thunis, A. Clappier and G. Pirovano

Contributors: E. Pisoni, C. Guerreiro, A. Monteiro, H. Dupont

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**FAIRMODE**

Forum for air quality modelling in Europe

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## 1. Introduction

Air pollution is one of the main causes of damages to human health in Europe, with an estimate of about 390 000 premature deaths per year in the EU28, as the result of exposure to fine particulate matter (PM<sub>2.5</sub>) only (EEA, 2018). One of the main challenges to improve this situation is to understand the origin of the pollution to make sure that air quality plans are targeting the appropriate sources at the right scale to ensure effective results. Source apportionment is used to face this challenge. In this document, we use a broad definition of source apportionment (Belis et al. 2019) to reflect the variety of usages currently covered by this discipline.

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**Source apportionment** is a technique used to relate emissions from various pollution sources to air pollution concentrations.

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Source apportionment can be applied to different pollutants. In the context of this guide, we address on the most critical pollutants: particulate matter, ozone and nitrogen dioxides, although the initial focus is on particulate matter. The source apportionment of ozone and nitrogen dioxide is discussed in the open issues section.

This document aims at supporting organizations in charge of air quality management in the context of the EU Ambient Air Quality Directives. In particular, it provides information on the different source apportionment approaches that are currently available, details their main characteristics and argue on their fitness-for-purpose. Finally, it also aims at supporting the interpretation of their results in the context of their applications.

In the context of the AAQD, source apportionment is used to support air quality planning. However, we also discuss in this document the use of source apportionment for the more general objective of supporting air quality management practices, in particular to improve the quality assurance of the overall modelling chain.

This guide is structured around four main chapters. We first review the main source apportionment methodologies and related concepts. The second chapter describes a theoretical illustrative example on which the concepts are applied while the third chapter addresses the aspect of fitness-for-purposes of the different approaches. Finally, open issues are discussed in the fourth section.

# Part I: Methods and Concepts

## 2. Which methods for source apportionment?

We distinguish in this document three main types of source apportionment results that we refer to as: (1) **impacts**, (2) **contributions** and (3) **increments**. Different type of results can be used in combination.

### 2.1. Impacts

Impacts can only be calculated with models, which can be of different types: Gaussian, Lagrangian, Eulerian or simplified source-receptor models based on any of these. The differences between these models are discussed in EEA (2011) or in Mircea et al. (2019). The method used to calculate impacts, often referred to as “brute-force”, “sensitivity analysis” or “perturbation method” is referred to as “**Emission Reduction Impact**” (ERI) in this document, which we refer to as “impacts” in the remaining of this document for convenience. The impact of a specific source is the difference between a model base case simulation (with full emissions) and a simulation in which the source emissions are reduced by a factor  $\alpha$ , divided by  $\alpha$  ( $Impact = \Delta C(\alpha)/\alpha$ ). An impact based on  $\alpha=20\%$  is then representative of moderate emission reductions (i.e. close to the baseline levels) while at  $\alpha=100\%$  (also referred to “zero-out”) the impact is representative of a complete activity switch-off. If chemical processes are non-linear for a given species, the two impacts differ. The method can be applied to any pollutant. We schematically represent the method to obtain impacts in Figure 1 below, where square symbols indicate that impacts are model based.

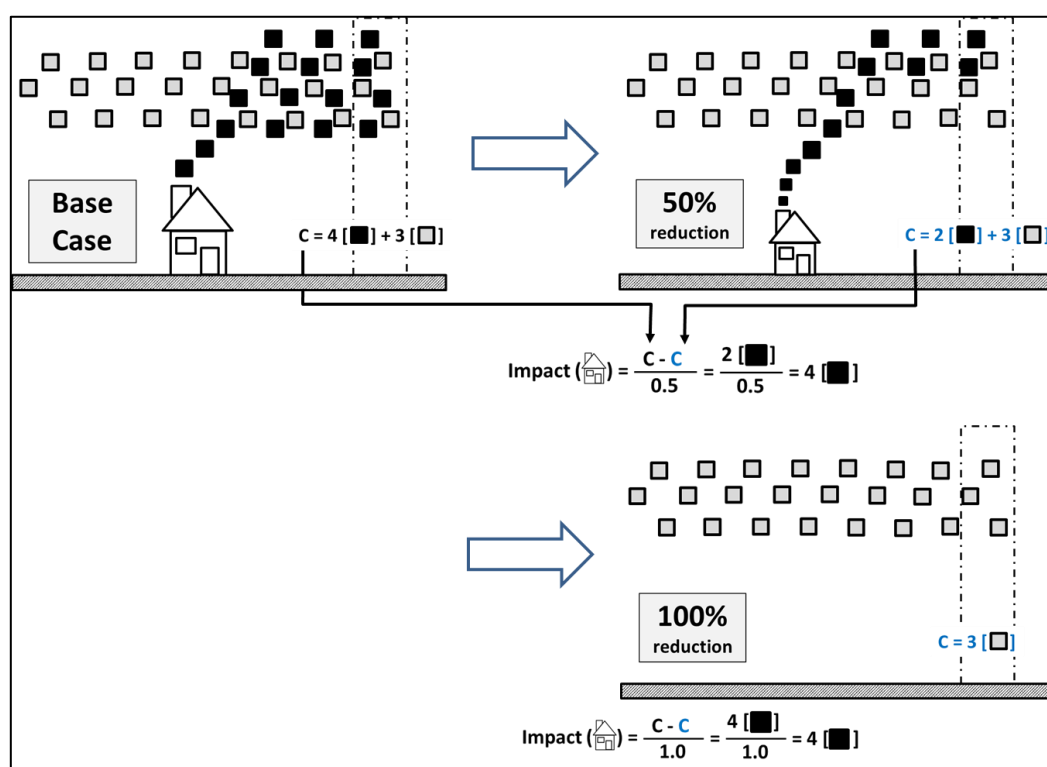


Figure 1: In this example, residential emissions (black squares) mix with the background pollution (grey squares) and lead to a given concentration downwind of the source (right dashed rectangle). When the source is reduced by 50% (right top), two out of the four black squares remain together with the background while for a full reduction, only the background remain (right bottom). Impacts correspond to the change of mass (projected to 100%) that results from the reduction or elimination

of the emission source, i.e. the difference between the downwind concentrations, with and without the source emissions, scaled by the percentage reduction: four black squares in our example. Squares are used to represent model-based output.

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**Impacts** correspond to the pollutant mass obtained by differencing two air quality model (AQM) simulations performed with the full emission source and a reduced emission source, scaled by the reduction intensity.

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## 2.2. Contributions

Contributions can be calculated starting from measurements (via receptor-oriented models) or starting from model results (source-oriented models using a tagging algorithm). The differences between these models are discussed in Mircea et al. (2019). Methods that deliver contributions are referred to as “**Mass Transfer**” (Thunis et al. 2019). For receptor models (Figure 2 – left), information on the type of emissions from the source is known and can be used to identify the contribution of the source in the final concentration, downwind of the source. This approach is based on measurements (solid circles) and is mostly applied to particulate matter.

For source-oriented models (Figure 2 – right), source precursors are tagged within an AQM. For non-linear components, the preserved atoms (e.g. Nitrogen, Sulphur ...) are tracked through the chemistry. The labels can be defined flexibly discriminating e.g. countries/provinces, sectors or fuel type. Hence, besides the concentration of each tracer also the corresponding fractional contribution of each label is calculated (Timmermans et al. 2017; Kranenburg et al. 2013). The method can be applied to any pollutant. Square symbols are used to indicate that these approaches are model-based. Since tagging contributions depend on the AQM, they therefore require all traditional AQM inputs to be available, in particular detailed emission inventories.

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**Contributions** correspond to the mass of a pollutant transferred from the emission sources to the ambient concentrations.

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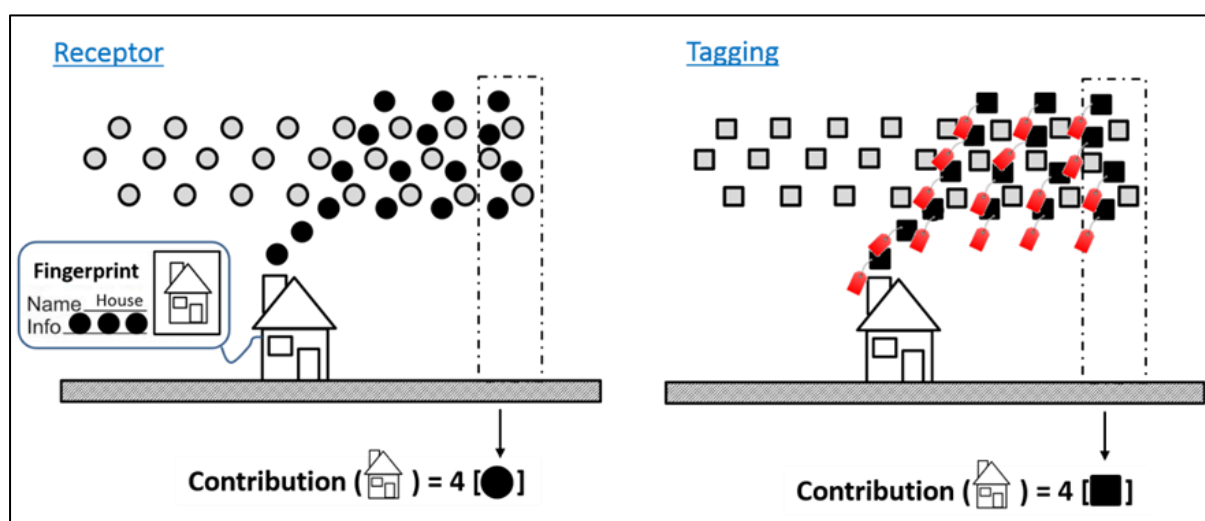


Figure 2: In this example, residential emissions (black symbols) mix with the background pollution (grey symbols) and lead to a given pollutant concentration downwind of the source (dashed rectangle). Contributions are obtained by (1 - left): recognizing in the downwind concentration (via pre-established sources emission fingerprints) the emitted pollutant from

the source or (2 – right) by tagging the emission precursors. Both options lead to four black symbols in our example. Circle and square symbols are used to differentiate measurement- from model-based approaches.

### 2.3. Increments

Increments are calculated using measurements. The method used to calculate increments, often referred to as “Lenschow” is referred to as “**Incremental**”. The method is generally applied to particulate matter but can be applied to any pollutant. We schematically represent the method to obtain increments in Figure 3 below.

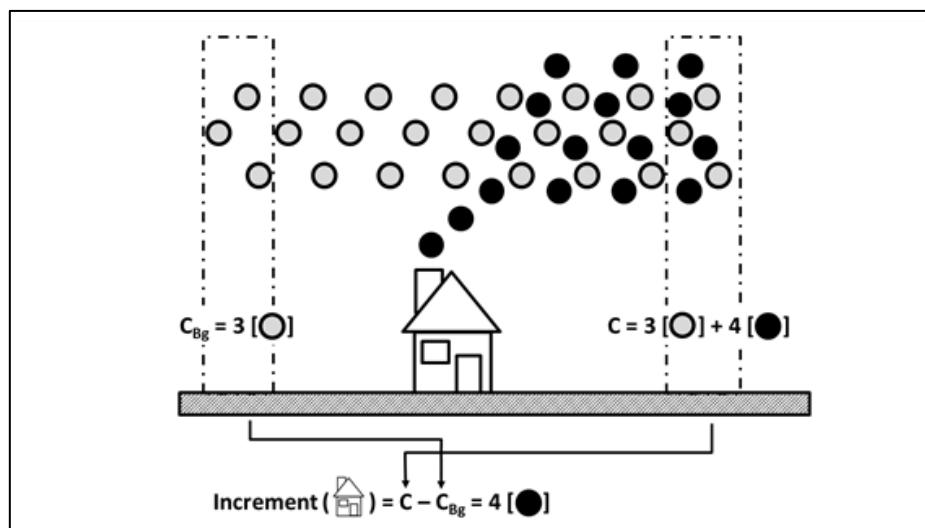


Figure 3: In this example, residential emissions (black circles) mix with the background pollution (grey circles) and lead to a given pollutant concentration downwind of the source (right dashed rectangle). Increments are obtained by subtracting the background concentration ( $C_b$ ) from the concentration downwind of the source, i.e. four black circles in our example. Circles are used as symbols in this figure because increments are mostly based on measurements.

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**Increments** are based on spatial gradients of concentration and are calculated as the difference between concentrations at two specific locations (one influenced by the source, the other not).

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Increments are most often measurement-based but can also be obtained via AQMs.

### 2.4. Combined methods

Source apportionment applications often use methods in combination. The Berlin air quality plan (Berlin2014) distinguishes the urban from the regional components with increments while impacts are used as a follow-up step to identify and quantify the sectoral origins of the pollution. The same approach is used in the case of Stockholm (Segersson et al. 2017).

Mertens et al. (2018) use impacts and contributions in complement, the first to assess the efficiency of mitigation measures on  $O_3$  levels and the second to retrieve additional information on unmitigated emission sources (i.e. those not covered by the impacts).

The combination of approaches is discussed further in the “open issues” section.

*In the above examples, all methods deliver the same results. One of the reasons is that only non-reactive compounds were considered. In real-world applications, this is often not the case and it is*

*therefore important to understand when, where and for which pollutant one method is suitable for a given purpose. In the next sections, we describe more complex situations and use more complete examples to illustrate these differences and highlight their implications.*

### 3. Types of atmospheric pollutants: linear vs. non-linear

Regardless of the source apportionment approaches used, it is important to distinguish species that behave linearly from those that do not. Linearity is a general property, which expresses the fact that an effect is proportional to a cause. For atmospheric pollutants, this translates in the definition below.

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**Linearity:** An atmospheric compound behaves linearly when the concentration change of that compound relates linearly to the strength of the emission sources.

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**Linear behaviour:** To illustrate this property, we use the example of two types of primary PM (PPM<sub>1</sub> and PPM<sub>2</sub>) that are emitted and mix in the atmosphere (Figure 4 – middle dashed rectangle). Since they do not interact between them, the measured PM compounds (right) correspond to the emitted ones. As shown in the scatter diagram, the concentration of PPM<sub>1</sub> (●) is directly proportional to the PPM<sub>1</sub> emissions (●) and does not depend on the PPM<sub>2</sub> emissions (●). PPM<sub>1</sub> behaves linearly.



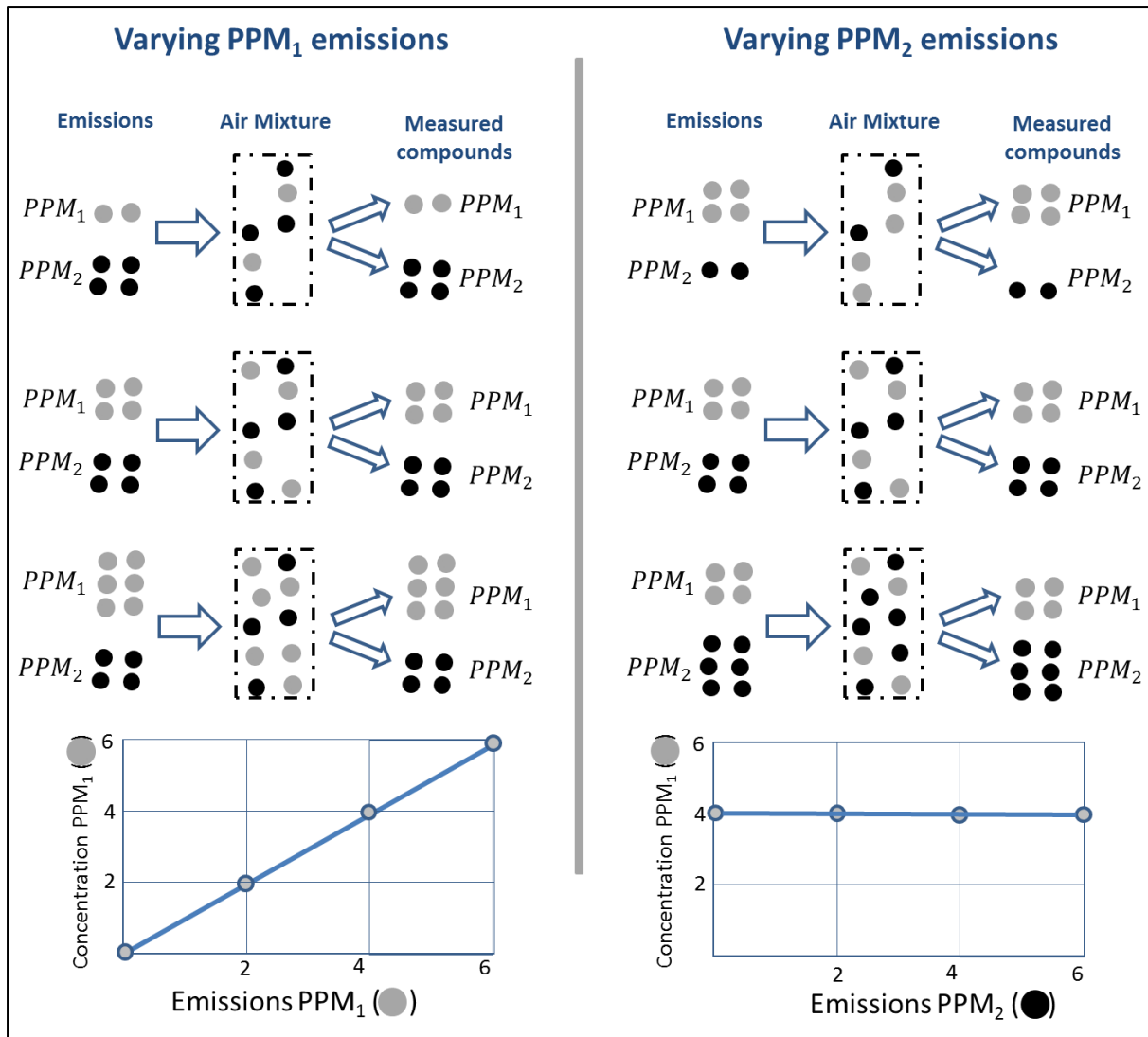


Figure 4: Schematic representation of a simple situation where two types of primary PM pollutants ( $PPM_1$  and  $PPM_2$ ) are emitted in the atmosphere and lead to specific downwind measurements. Because they do not interact between themselves, the emitted pollutant are the same as those measured. Situations corresponding to different share of emissions are shown for  $PPM_1$  (left) and  $PPM_2$  (right). The two scatter diagrams illustrate how the  $PPM_1$  concentration varies with the strength of the emissions of  $PPM_1$  (left) and  $PPM_2$  (right).  $PPM_1$  varies linearly with  $PPM_1$  emissions. The same holds for  $PPM_2$ .

**Non-linear behaviour:** In our second example, two gas-phase precursors:  $NO_x$  (○) and  $NH_3$  (○) combine on a 1:1 basis to create ammonium nitrate ( $NO_3NH_4$ ):  $\text{○} + \text{○} \rightarrow \text{●}$ . Out of ammonium nitrate (●), nitrate ( $NO_3$  - ●) and ammonium ( $NH_4$  - ●) are the measured compounds (Figure 5). If we focus on nitrate (similar conclusion can be made for ammonium), we see that its concentration varies with the emission strengths of both  $NH_3$  and  $NO_x$ , in both cases in a non-linear manner. Nitrate behaves therefore as a non-linear compound.

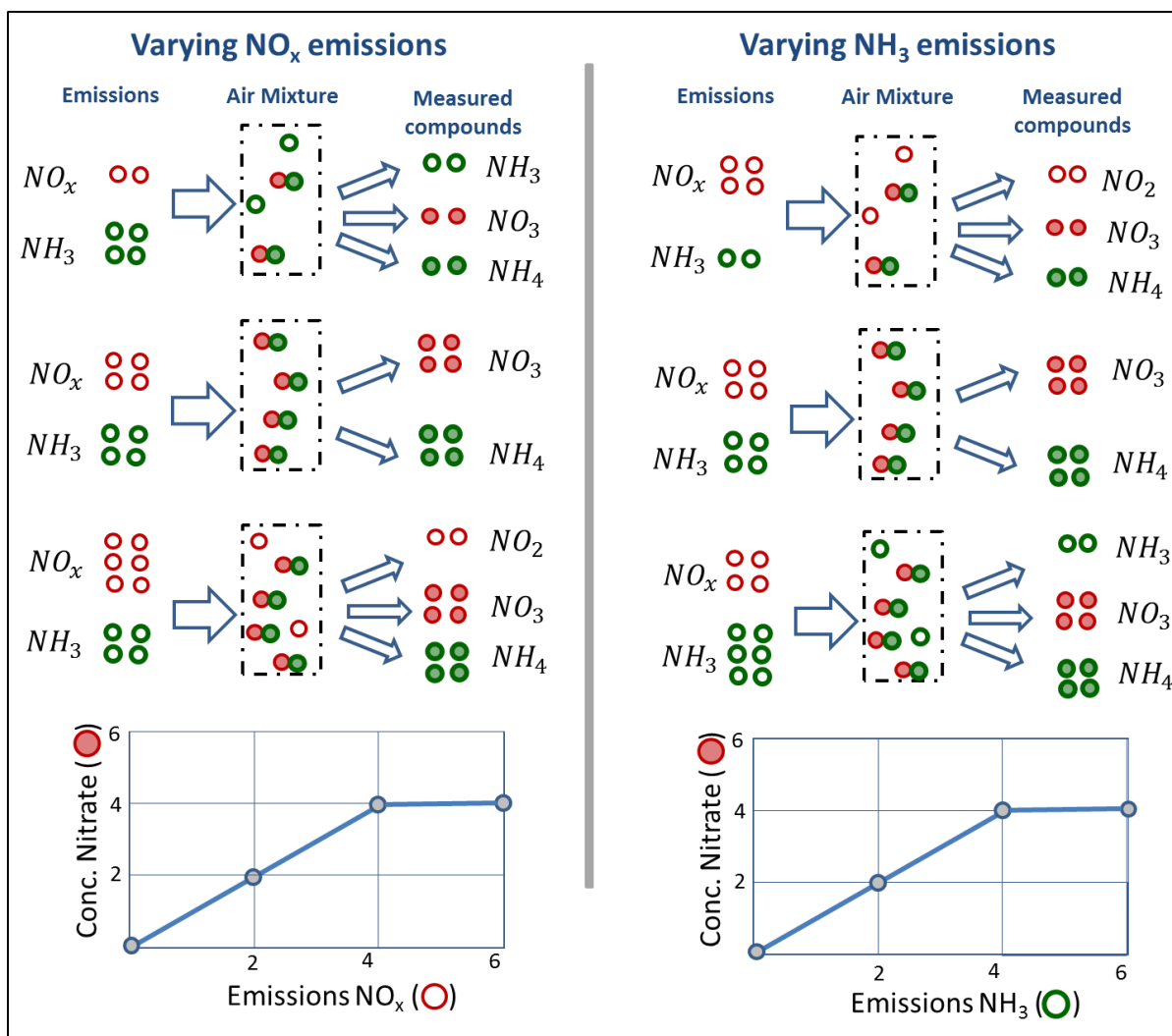


Figure 5: Schematic representation of a simple situation where two types of gas-phase precursors: NO<sub>x</sub> (○) and NH<sub>3</sub> (●) are emitted in the atmosphere, react on a 1:1 basis to form ammonium nitrate (NH<sub>4</sub>NO<sub>3</sub> - ●). The atmospheric mix is then decomposed in the different measured compounds: NO<sub>x</sub>, NH<sub>3</sub>, nitrate (NO<sub>3</sub> - ●) and ammonium (NH<sub>4</sub> - ●). Because they do interact between themselves, the measured pollutants can be different from those measured. Situations corresponding to different share of emissions are shown for NO<sub>x</sub> (left) and NH<sub>3</sub> (right). The two scatter diagrams illustrate how the nitrate concentration (NO<sub>3</sub>) varies with the strength of the emissions of NO<sub>x</sub> (left) and NH<sub>3</sub> (right). Nitrate varies non-linearly in terms of both the NO<sub>x</sub> and NH<sub>3</sub> emissions. A similar behaviour (not shown) would be obtained for ammonium (NH<sub>4</sub>).

Because the results of different source-apportionment methods vary when we apply them for linear or non-linear chemical species (because of their intrinsic assumptions), we distinguish linear from non-linear species in our analysis. While for some species, it is straightforward to assess a linear or non-linear behaviour, this is not always the case. This is discussed in the “open issues” section.

**Direct/non-direct effects:** In the example presented above, secondary products (NO<sub>3</sub>NH<sub>4</sub>) result from the interaction between different precursor emissions (NO<sub>x</sub> and NH<sub>3</sub> in our case). This interaction is reflected by the fact that nitrate (NO<sub>3</sub>) – a similar reasoning can be made for ammonium (NH<sub>4</sub>) – depend on NO<sub>2</sub> but also on NH<sub>3</sub> emissions. In literature, the dependence of NO<sub>3</sub> to its direct precursor NO<sub>2</sub> is often referred to as a *direct effect* while the dependence of NO<sub>3</sub> to NH<sub>3</sub> is referred to as the *indirect effect*.

Because of this secondary products interaction, it is challenging for source apportionment to determine which fraction of ammonium nitrate (NO<sub>3</sub>NH<sub>4</sub>) (or nitrate or ammonium) originates from

the  $\text{NO}_2$  emissions on one side and which fraction originates from the  $\text{NH}_3$  emissions on the other. In tagging/labelling approaches this issue is solved by neglecting the non-direct effects ( $\text{NO}_3 = f(\text{NO}_2)$  and  $\text{NH}_4 = f(\text{NH}_3)$ ). In other words, ● is attached to ○ and ● is only attached to ○ (see application in Section 5.2).

#### 4. Which are the properties of source apportionment approaches?

Before proceeding with more complex examples, we first discuss shortly in this section a few concepts to support our analysis. We discuss in particular a few properties that can be associated to source apportionment approaches. These properties will then be helpful to discuss their fitness-for-purpose. As done so far, we will continue to use circles and squares symbols to differentiate measurement- from model-based approaches.

##### 4.1. Measurement vs. Model-based

As highlighted, source apportionment methods can be measurement- or model-based or accommodate both options. While the incremental and receptor approaches are mostly based on measurements, impacts and tagging contribution can only be obtained from models.

##### 4.2. Unambiguity

**Unambiguity:** A source apportionment approach is unambiguous when each component relates explicitly to one and only one source.

To illustrate this property, we apply the incremental approach to two situations (Figure 6). In the first case (left), the background (empty circles) is constant everywhere and the source only influences the downwind location. In the second case (right), none of these two conditions is fulfilled. Although different symbols are used for the background and residential pollution, both the empty and solid circles represent the same compound in these two cases. While the source and background increments are unambiguously related to their original precursors in the first situation, this is not the case in the second as both increments mix precursors from both sources (● and ○).

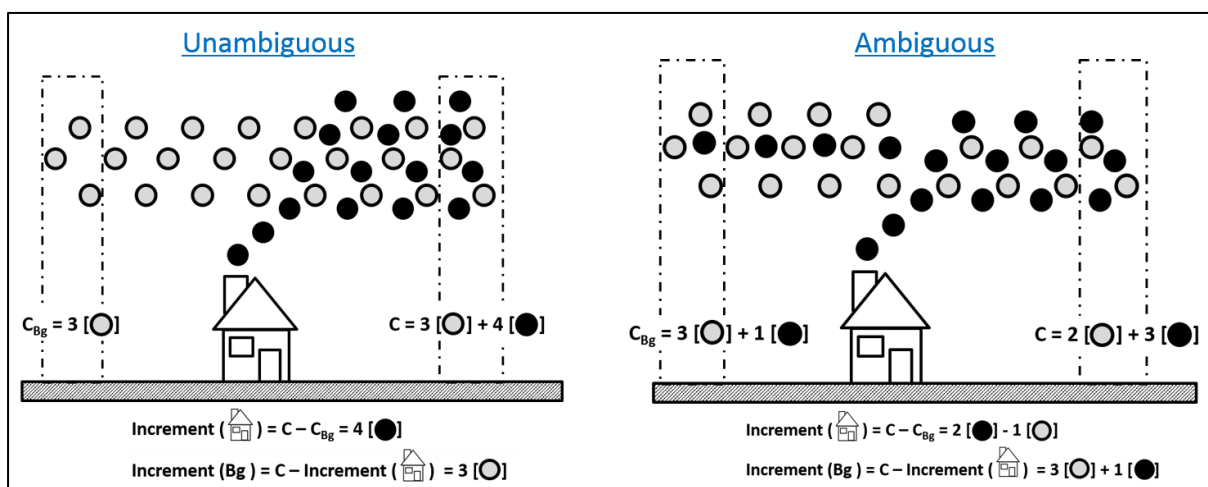


Figure 6: Incremental approach applied to two different situations (See text for details).

The two conditions mentioned above (constant background and no influence of the source on the background) are the two conditions underpinning the incremental approach. These two conditions are developed in Annex A.

In this example, ambiguity appears because measurements are not able to distinguish between the pollutants emitted by the source and those coming from the background. Note that a Mass Transfer method based on receptor models or tagging, or an impact based approach would remove the ambiguity.

### 4.3. Additivity

**Additivity:** The sum of the individual source apportionment components (C) equals the combined (all sources at once) component. In other words, for two sources A and B:  
 $C_{AB} = C_A + C_B$ .

To illustrate this property, we apply the impact approach to the same situation as previously analyzed with the addition of a constant background (Figure 7). The residential impact on (C) is obtained by switching off the residential emissions (top right) and differencing with the base case. The industrial impact is obtained similarly by switching off the industry emissions while the combined impact is obtained by switching off both sources contemporaneously.

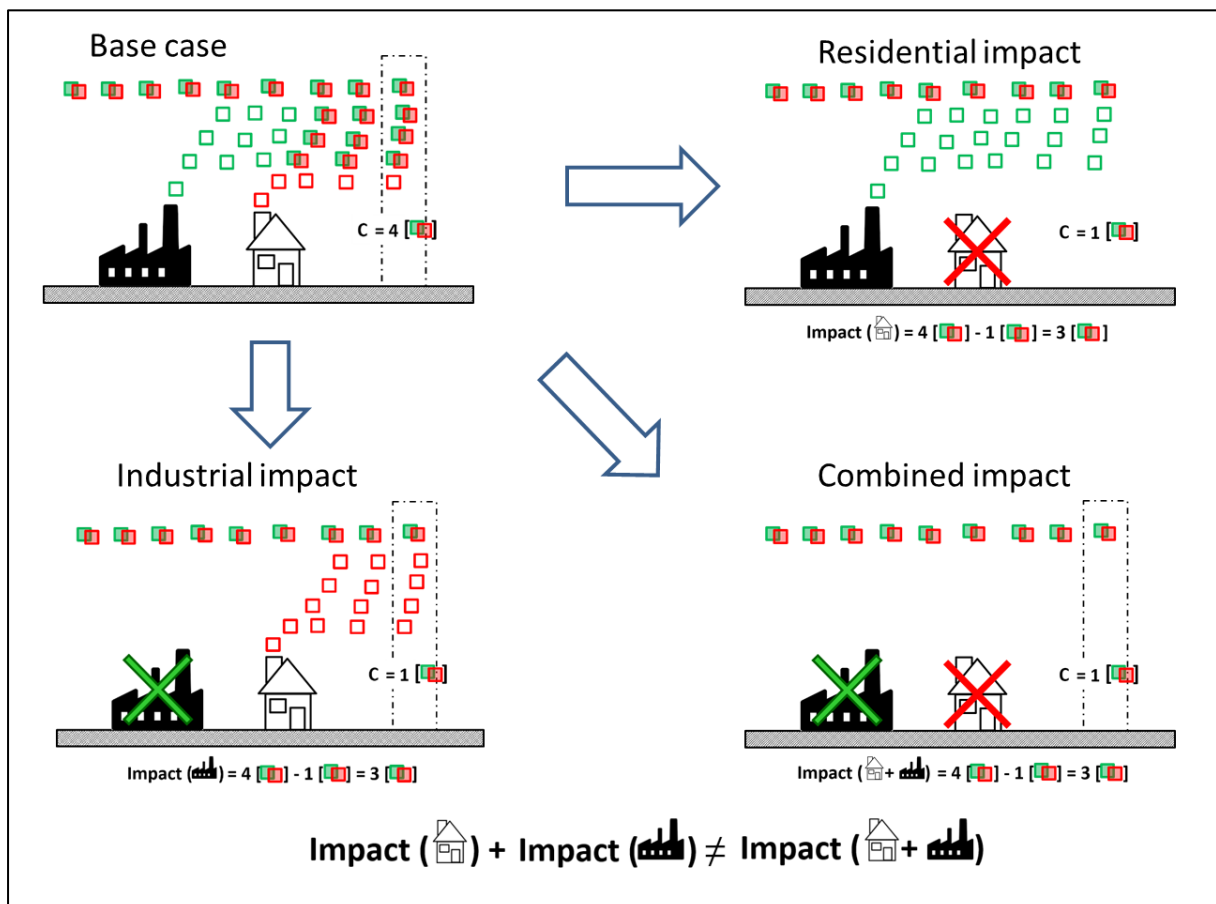


Figure 7: Assessment of the additivity. From the base case situation (top left), the industrial impact is calculated by subtracting from the base case the concentration obtained when industry is switched off (bottom left) while the residential impact is obtained similarly with the residential emissions (top right). The combined impact is obtained by switching off

both sources (bottom right). We use here square symbols (rather than circles) to highlight the fact that impacts are model-based.

This example illustrates a case for which additivity is not fulfilled. It would be possible to reach an equality by introducing an additional term, equal to the difference between the sum of the single source impacts and the combined (both sources) impact. This additional term is called interaction term in literature (Stein and Alpert, 1993) because it represents interaction between the sources. As it represents an interaction between sources, this term is however ambiguous.

#### 4.4. Linearity




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**Linearity:** A source apportionment method is linear when the output signal strength varies in direct proportion to the input signal strength. In our case, this means that the SA component is proportional to the source strength.

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To illustrate this property, we compare the mass-transfer (tagging) and Emission reduction impacts approaches and assess whether their outcome (contributions and impacts) are proportional to the source strength.

For the impacts, (Figure 8 – left column), different reduction percentages are applied to the residential emissions while the background is kept constant. The impact (difference between the scenario and the base case) obtained for each percentage is reported below each figure as well as in the scatter diagram. Impacts are not linear with respect to the strength of the residential emissions.

For contributions, (Figure 8 – right column), the source strength is changed by tagging only a percentage of its emissions. As mentioned in Section 3, each PM component in tagging/labeling approach is linked only to its direct primary precursor (e.g.,  $\text{NO}_3 = f(\text{NO}_2)$  and  $\text{NH}_4 = f(\text{NH}_3)$ ). Nitrate ( $\text{NO}_3$  - ) is therefore attributed to residential because only the residential sector emits  $\text{NO}_x$  () and  $\text{NH}_4$  () is attributed to the background. The contribution (sum of red tagged PM) obtained for each tagged percentage is reported below each figure as well as in the scatter diagram. Contributions are linear with respect to the strength of the residential emissions.

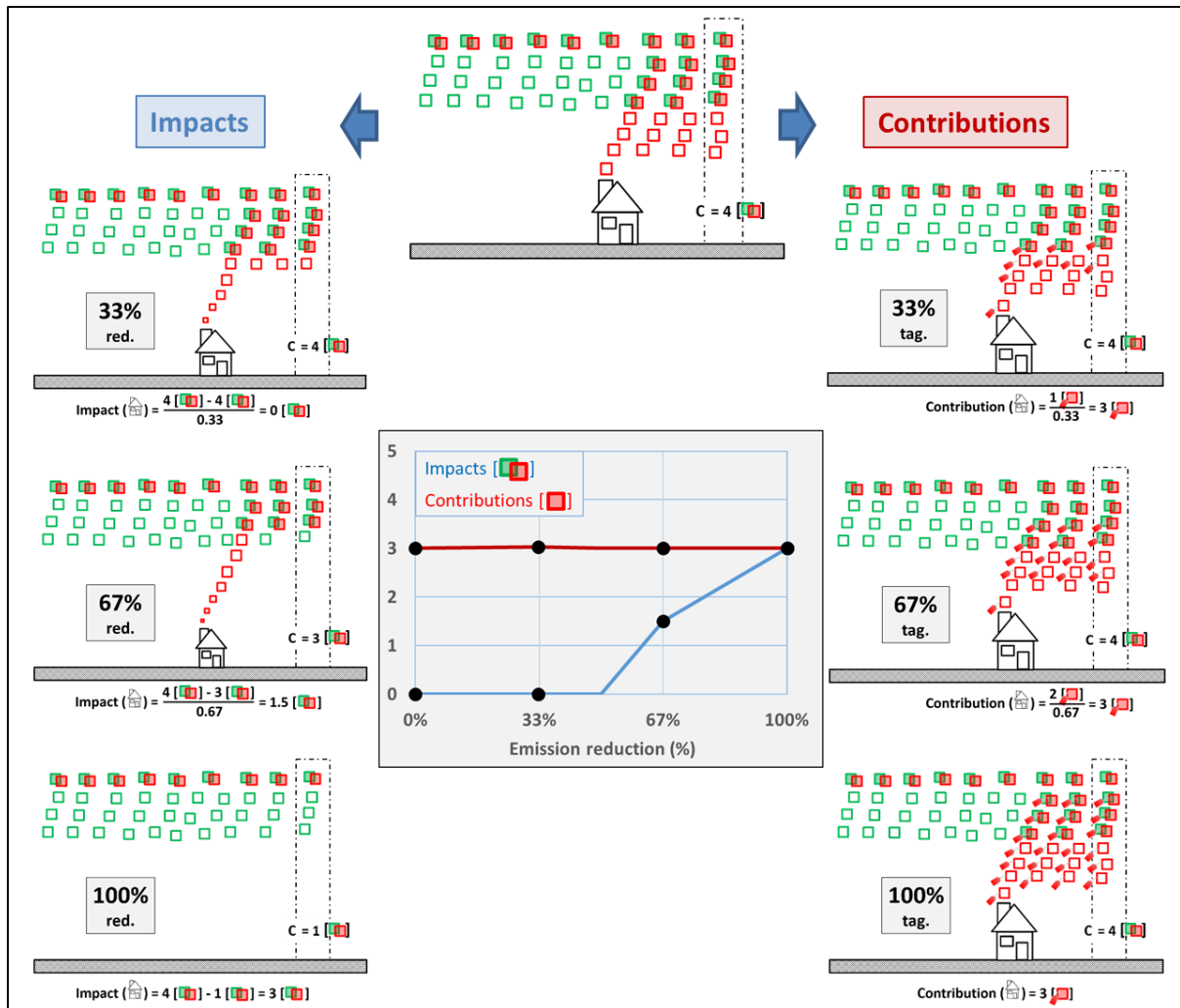


Figure 8: Example of non-linear relationship between emission and concentration. Two sources (industrial and residential) emit gas-phase precursors (green and red empty squares, respectively) which combine in particulate matter (mixed shaded squares) measured at a given location (dashed rectangle). On the left side, the strength of the residential source is reduced by 33, 67 and 100% to obtain the impacts on PM concentrations. Similarly, on the right side, only a fraction of the residential source emissions (33, 67 and 100%) is tagged to obtain the corresponding contributions. The impacts and contributions obtained for the different percentages are graphically represented in the scatter diagram. In this figure, the empty green squares (□) are the modelled  $\text{NH}_3$ , the red empty squares (□) are the modelled  $\text{NO}_2$ , the shaded green square (■) ammonium, the shaded red square (■) the modelled nitrate and the mixed green-red shaded binome (■) the ammonium nitrate.

## 4.5. Dynamicity

**Dynamicity:** A source apportionment approach is dynamic when its components reflect the influence of emission changes on concentration.

To illustrate this property, we use the same example as presented in the previous Section for the specific conditions of 100% (Figure 8). The reference in this case is the impact because it is constructed to reflect the consequence of an emission reduction on concentration. As seen from this example the tagging contribution (3 ■) is not similar to the impact (3 ■) because the masses of  $\text{NO}_3$  (■) and  $\text{NO}_3\text{NH}_4$  (■) are different. The contributions are therefore not dynamic while the impacts are.

#### 4.6. Consequences on the interpretation of SA results

Source apportionment results are usually reported in terms of a pie chart in which the various sources are expressed as a percentage of the total mass (Figure 9).

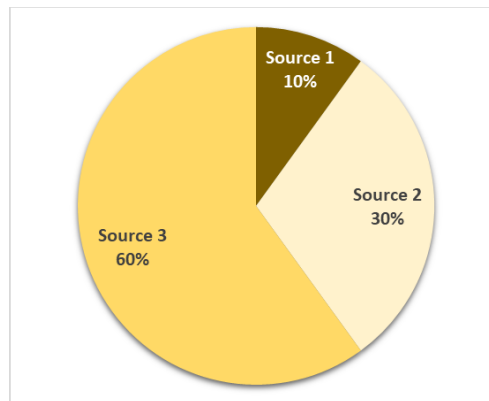


Figure 9: Example of pie chart showing the percentage share of three sources to the total concentration

While pie charts provide a straightforward way to present results, their interpretation can be misleading when some of the properties detailed above are not fulfilled. We list below some potential issues.

Lack of additivity: If additivity is not guaranteed, source impacts cannot be summed. For example, the impact of switching off both sources 1 & 3 will not equal the sum of the individual impacts as indicated in the pie-chart (i.e. impact (1&2)  $\neq$  10%+30%). A general issue is also that the sum of the three sources will not be equal to the total modelled/measured mass. It then become impossible to represent the SA results via a pie-chart.

Lack of linearity: If linearity is not guaranteed, it is not possible to extrapolate the pie-chart values to other source strengths than those on which the pie chart is based. For example, the contribution of half of source 1 will not be equal to  $10/2=5\%$ .

Lack of dynamicity: If the SA approach is not dynamic, pie chart values cannot be used to inform on the mass decrease that would result from an emission reduction. For example, the contribution of source 2 (30%) cannot be used to tell how much the concentration would change when emissions from this source are reduced by any percentage. This is only possible with impacts-based approaches, under certain conditions (see next Section).

When the source apportionment method is additive, linear and dynamic the pie chart is extremely easy to use to estimate the impact of an emission reduction. We just have to multiply the percentage of emission reduction by the percentage share of the source to be reduced. For example, the impact of a 50% reduction of source 2 will be equal to  $50\% \times 30\% = 15\%$ .

#### 4.7. Linear and additive impacts: the case of source allocation

The impact of a source can be estimated by switching off emissions entirely ( $\alpha = 100\%$ ) for a given sector/area as in the example illustrated in Figure 1. The method is then referred to as “zero-out” (Osada et al. 2009; Wang et al. 2014; Wang et al. 2015; Huang et al. 2018;). The impact of a source can also be estimated by reducing emissions by a smaller amount and scale the concentration change to 100% (e.g. multiply by five the concentration change resulting from a 20% emission reduction, assuming a linear behaviour). Methods using lower emission reductions (e.g. 20% as in

Koo et al., 2009; 15% as in EMEP or 50% in SHERPA (Thunis et al. 2018) that conserve linearity and additivity are referred to as source allocation. This is explained in Figure 10 and Figure 11.

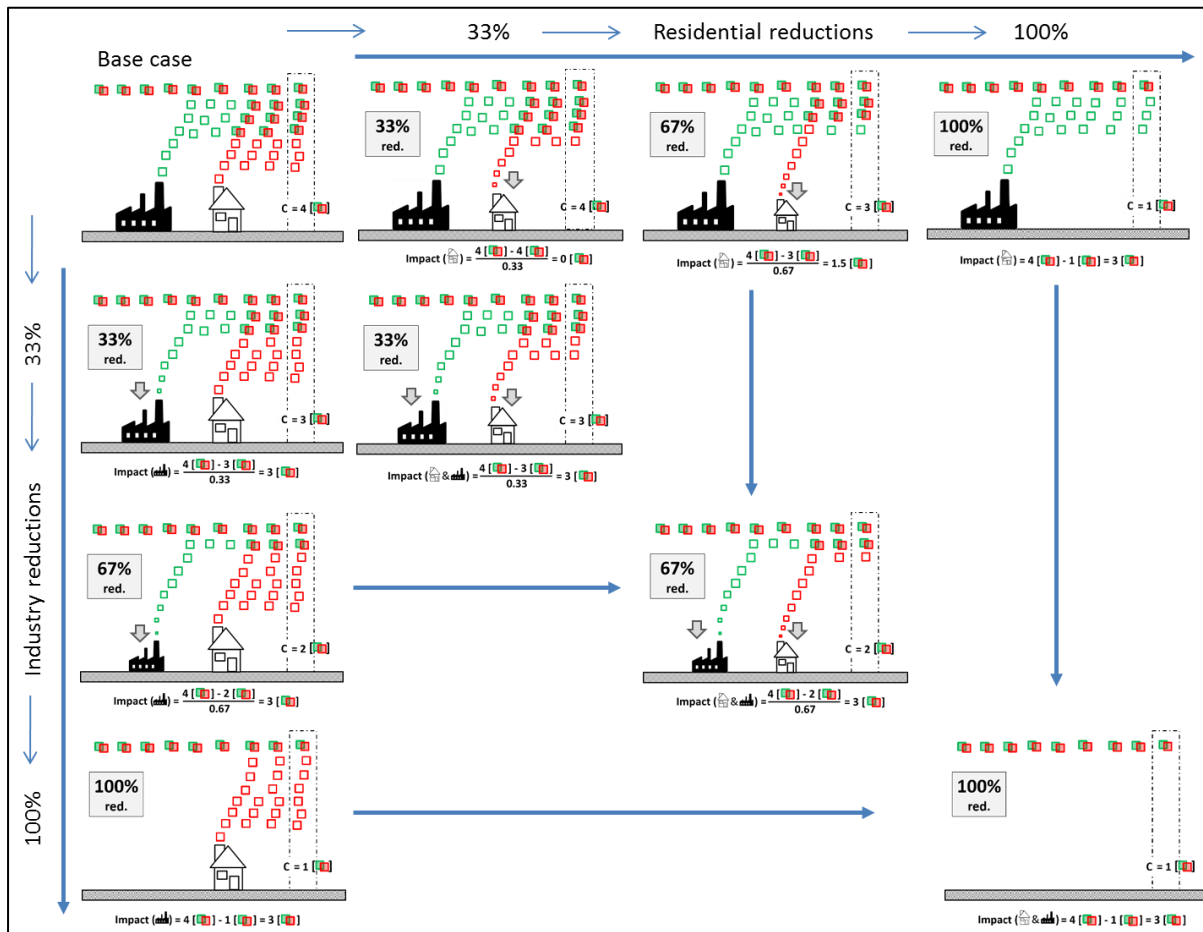


Figure 10: Impacts obtained for different emission reduction levels (33, 67 and 100%). The impacts are obtained by differencing the base case results (top left figure) and each of the emission reduction scenarios: the scenarios include reduction of the residential sources only (top row), of the industrial sources only (left column) and of both sources (central). A grey arrow is used to point to the reduced source.

The results obtained over the entire range of emission reductions (Figure 10) can be used to define the range of applicability of source allocation in terms of linearity and additivity (Figure 11).



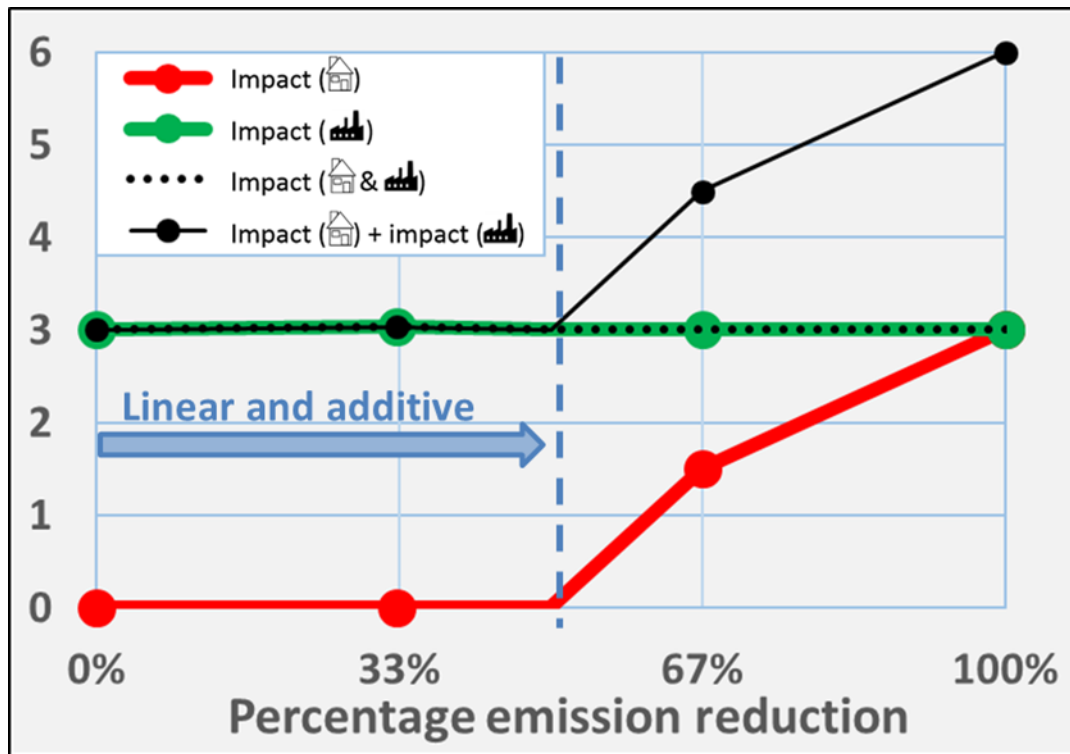


Figure 11: Based on the results of Figure 10, this diagram indicates how the residential (red) and industrial (green) impacts compare for different emission reduction values (X-axis). The dashed and solid lines represent the combined impact (both sources reduced simultaneously) and the sum of the two single impacts, respectively. The blue dashed line shows the application limit of the source allocation approach (left side of the diagram) that preserves linearity and additivity.

## Part II: An illustrative (theoretical) example

In this section, we use a more complete example to illustrate how the components obtained with different approaches compare in terms of their sectorial (section/chapter 5) and spatial apportionments (section/chapter 6). We assess in particular how the different components compare in terms of the characteristics addressed in the previous section.

### 5. Sectorial apportionment

Our simple theoretical example (Figure 12) consists of two sources (industry and residential) emitting three different pollutants, PPM (black solid circles),  $\text{NO}_x$  (red empty circles) and  $\text{NH}_3$  (green empty circles) into a constant background composed of secondary ammonium nitrate (combined green-red shaded circles), PPM and  $\text{NH}_3$ . The background PPM originates from different sources (residential, transport or dust). As dust differs from the other PPM emitted by transport or residential, we represent it with another colour (grey). While PPM remains passive, one mole of  $\text{NH}_3$  reacts with one mole of  $\text{NO}_x$  to generate ammonium nitrate ( $\text{NH}_4\text{NO}_3$ ), a combination of nitrate  $\text{NO}_3$  and ammonium  $\text{NH}_4$ . We only focus on particulate matter concentrations (i.e. the shaded symbols).

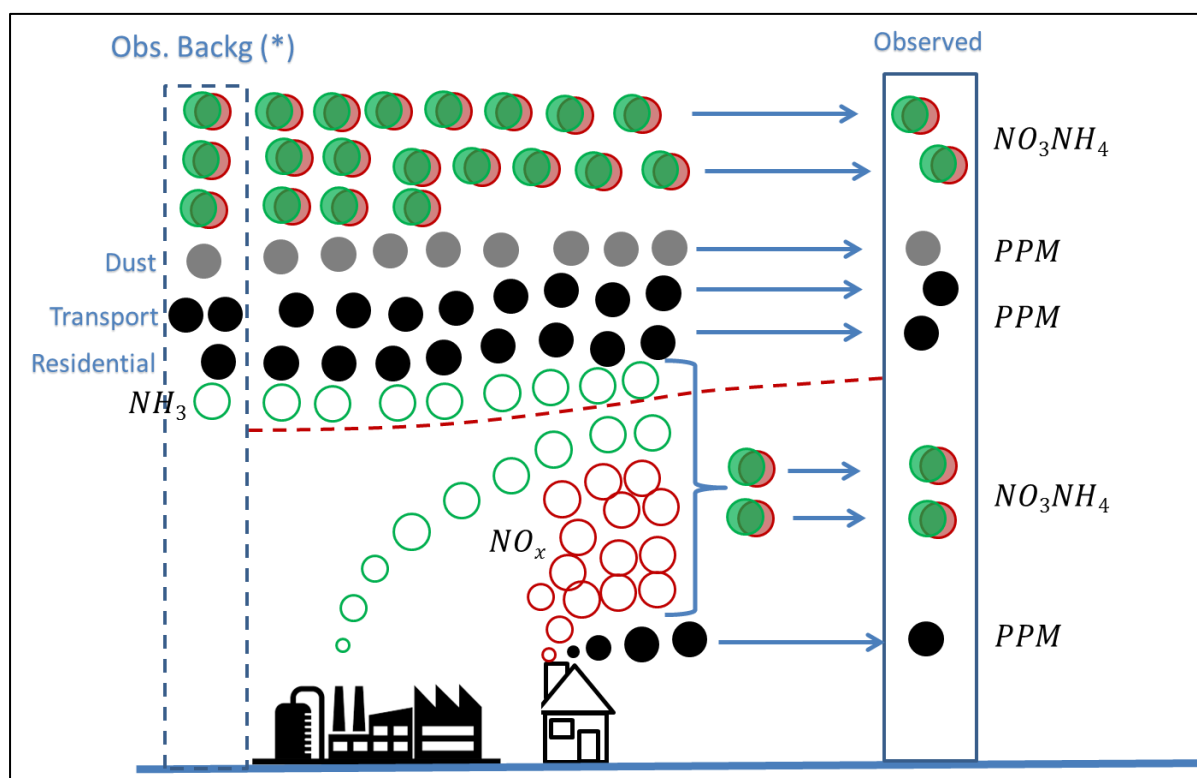


Figure 12: Illustrative theoretical example used to highlight differences between source-apportionment methods. The background pollution is distinguished from the local one by the red dashed line. The gas-phase compounds are not considered in the calculation of the final concentrations. See additional details in the text.

#### 5.1. Receptor contribution

From the observed concentration (left column in Figure 13), receptor contributions are obtained according to the following rules:

- For compounds emitted by a source that has a single spatial and sectorial origin (dust in our example), the apportionment is direct and can be made from measurement prior to the application of the receptor model (pink shading - point 1 in Figure).

- Receptor models apportion the mass of an atmospheric pollutant based on measurements. We highlight this important point by keeping circle symbols for each contribution.
- Because of their underlying assumptions, receptor contributions are limited to the apportionment of the linear fraction of the mass (middle column). In our example, this implies that nitrate, sulphate and other secondary components are not apportioned (red arrows leading to shaded rectangles on the final apportionment).
- Receptor contributions distinguish the sectoral origins of a similar compound emitted by different sources (difference between background traffic and residential – arrows (2)).
- Receptor contributions do not differentiate between background and local (arrows 2 & 3). The residential contribution represents therefore a mix between background and local origins.

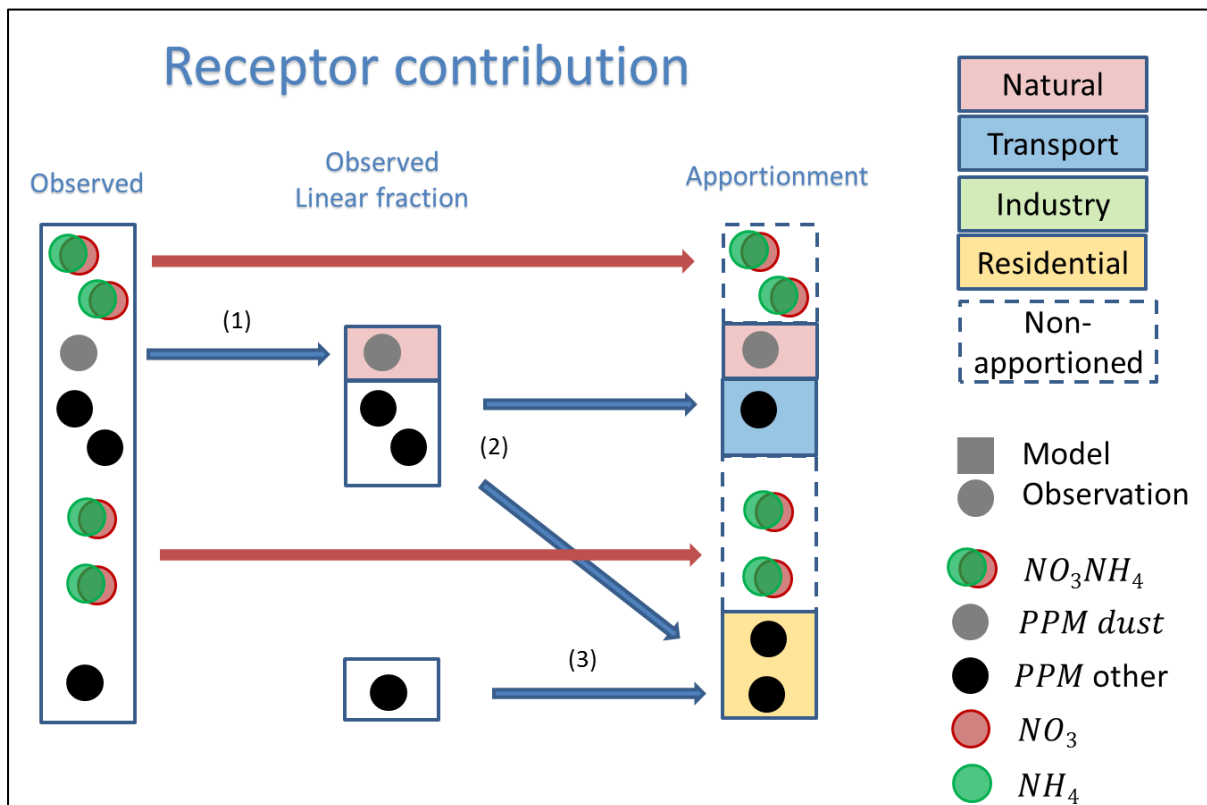


Figure 13: Process to obtain the receptor contributions, starting from the measurements (left column) towards the final sectoral contributions (right column). Sources that can be apportioned in terms of their sectoral origins are shaded with colors indicated in the top-right legend. Receptor contributions are measurement-based (circles) and only apportion linear species (middle column) and non-linear compounds are not apportioned (red arrows leading to shaded rectangles). Receptor models are able to distinguish the sectoral origins of a similar compound emitted by different sources (arrow 2) but cannot differentiate between background and local (arrow 2 & 3). Dust is apportioned directly from measurements as it originates from a single spatial and sectoral source.

## 5.2. Tagging contribution

From the observed concentration (left column in Figure 14), tagging contributions are obtained according to the following rules:

- Tagging contributions are based on a model as indicated by square symbols in the middle column.

- Compounds emitted by a source that has a single spatial and sectoral origin (dust in our example), the apportionment is direct and corresponds to the base case concentration for that compound. (pink shading - point 1 in Figure).
- With the exception of the source mentioned above (dust), only the sources emitted within the AQM modelling domain are tagged in terms of sectors and contributions from background species are therefore not apportioned sectorally (red arrows leading to shaded rectangles). The local sources are tagged and distinguished from the background.
- For secondary products ( $\text{NO}_3\text{NH}_4$  in our example) that results from the combination of local and background precursors, usually emitted by different sectors, the difficulty is to determine which fraction originates from a specific sector (e.g. residential). In tagging/labelling approaches, only the direct effects are considered (see Section 3). Nitrate ( $\text{NO}_3$ ) in the final secondary pollutant is therefore attributed to residential activity because only this sector emits  $\text{NO}_x$  while  $\text{NH}_4$  is attributed to both the industry and background.

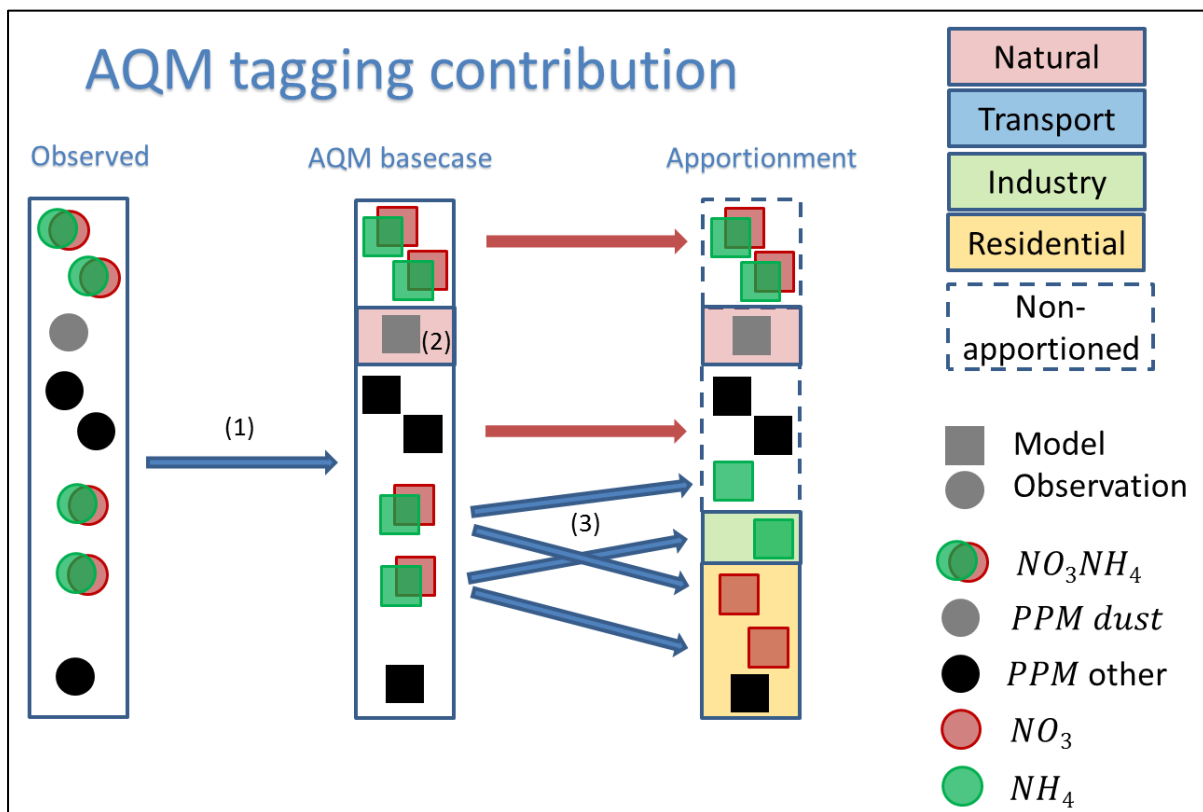


Figure 14: Process to obtain the tagging contributions, starting from the measurements (left column) towards the final sectoral apportionment (right column). Sources that can be apportioned in terms of their sectoral origins are shaded with colours indicated in the top-right legend. Tagging contributions are model-based (middle column - squares) and do not apportion the species that are emitted outside the modelling domain (red arrows leading to dashed contoured rectangles). Tagging contributions neglect the non-direct effects (only the red secondary fraction of the source is kept in the contribution - arrow 3). Dust is apportioned directly from the modelled base case concentration as it originates from a single spatial and sectoral source.

## 5.3. Impacts

### 5.3.1. Zero-out

From the observed concentration (left column in Figure 15), zero-out impacts are obtained according to the following rules:

- Impacts are model-based (square symbols, 2<sup>nd</sup> column).
- For compounds emitted by a source that has a single spatial and sectoral origin (dust in our example), the apportionment is direct and is equal to the base case concentration for that compound (pink shading - points 1 in Figure).
- The local industry and residential impacts are obtained as the difference between the modelled base case (column 2) and scenarios in which the local industry (column 3) and residential (column 4) sectors are switched off. Because only the sources emitted within the AQM modelling domain are reduced, background species are not apportioned (red arrows).
- For secondary products ( $\text{NO}_3\text{NH}_4$ ), impacts include an indirect effect which means that a reduction of the  $\text{NO}_x$  emissions has an influence on total ammonium nitrate, i.e. not only  $\text{NO}_3$  as for contributions but also  $\text{NH}_4$  (arrow (3))
- Given the large emission reductions applied, the sum of the impacts and non-apportioned fractions exceed the base-case AQM base-case concentration (5 vs. 4 moles of ammonium nitrate).

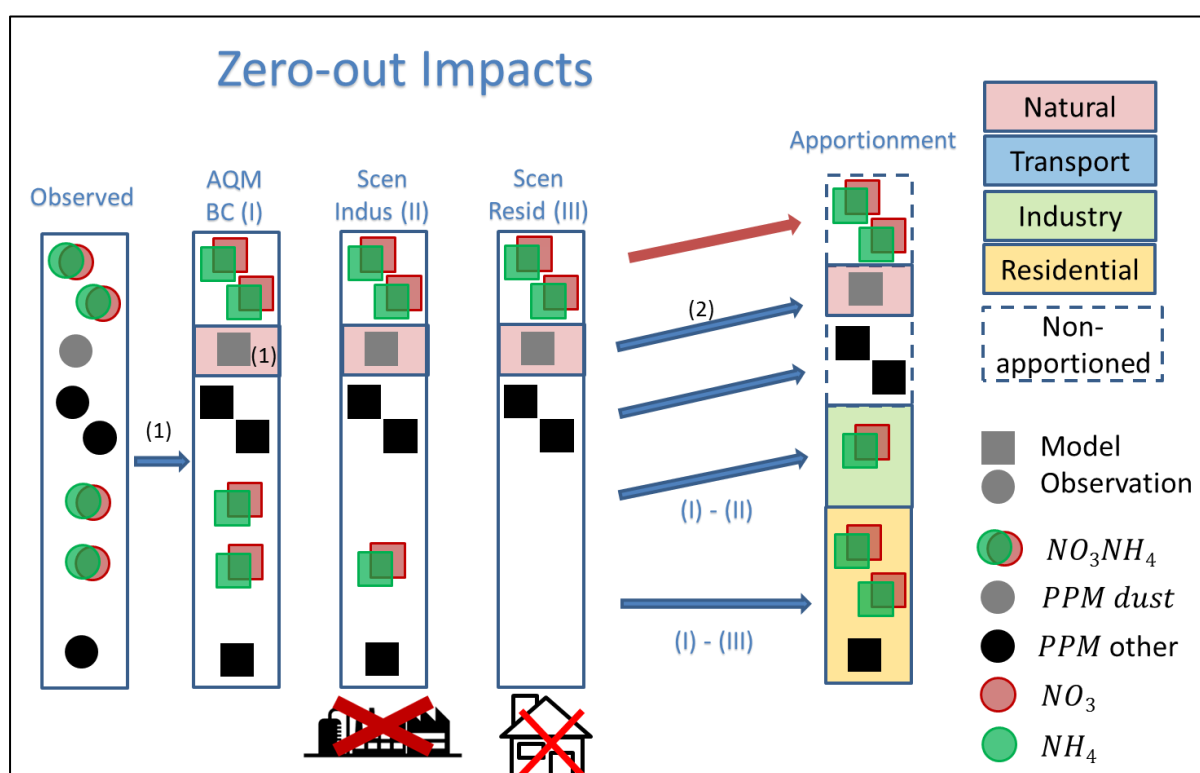


Figure 15: Process to obtain zero-out impacts, starting from the measurements (left column) towards the final sectoral apportionment (right column). Sources that can be apportioned in terms of their sectoral origins are shaded with colours indicated in the top-right legend. Impacts are model-based (squares in 2<sup>nd</sup> column and following) and are obtained as the difference between a model base case (column 2) and a scenario (column 3) in which the source is removed. Impacts only apportion the species that are emitted within the modelling domain (2) and include non-direct effects (3). Dust is apportioned directly from the modelled base case concentration as it originates from a single spatial and sectoral source.

### 5.3.2. Source allocation

From the observed concentration (left column in Figure 16), Source-allocation (SAL) impacts are obtained according to the following rules:

- Impacts are model-based (square symbols, 2<sup>nd</sup> column).
- For compounds emitted by a source that has a single spatial and sectoral origin (dust in our example), the apportionment is direct and can be made from the base case concentration prior to the application of the impact approach (pink shading - points 1 in Figure).
- The local industry and residential impacts are obtained as the differences between the modelled base case (column 2) and scenarios in which the local industry (column 3) and local residential (column 4) sectors are reduced. Given the 50% emission reduction applied, the impacts are obtained by multiplying these differences by a factor 2. Background species are not apportioned (red arrows).

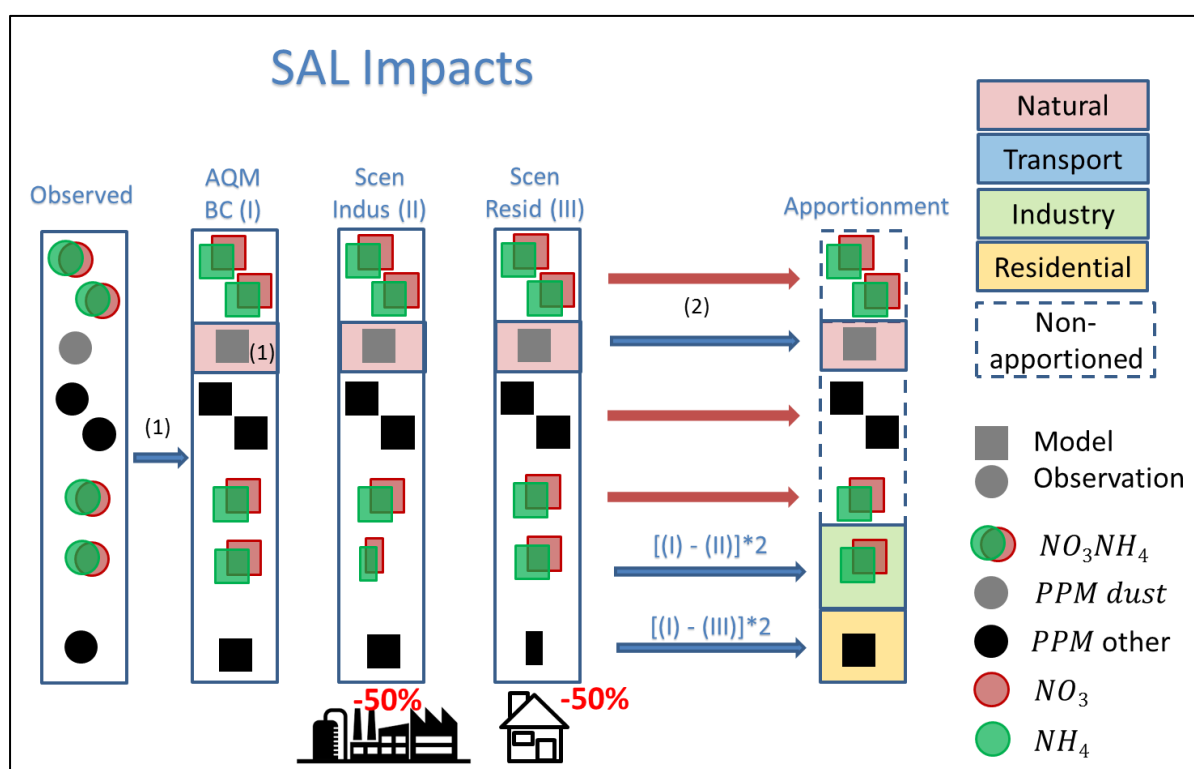


Figure 16: Process to obtain source allocation impacts, starting from the measurements (left column) towards the final sectoral apportionment (right column). Sources that can be apportioned in terms of their sectoral origins are shaded with colours indicated in the top-right legend. Source allocation impacts are model-based (squares in 2<sup>nd</sup> column and following) and are obtained as the difference between a model base case (column 2) and scenarios (columns 3 & 4) in which the sources are reduced (here by 50%). Source allocation Impacts do not apportion the species that are emitted outside the modelling domain (red arrows) and include non-direct effects. Dust is apportioned directly from the modelled base case concentration as it originates from a single spatial and sectoral source

### 5.4. Increments

From the observed concentration (left column in Figure 17), increments are obtained according to the following rules:

- For compounds emitted by a source that has a single spatial and sectoral origin (dust in our example), the apportionment is direct and can be made from measurement
- The background increment is equal to the measured background (column 2 from Figure 12).

- The local increment (3<sup>rd</sup> column) is obtained by subtracting the background increment from the observed concentration (1<sup>st</sup> column).
- From a sectoral perspective, the incremental approach does not deliver any information (right column), with the exception of dust (see first point above).

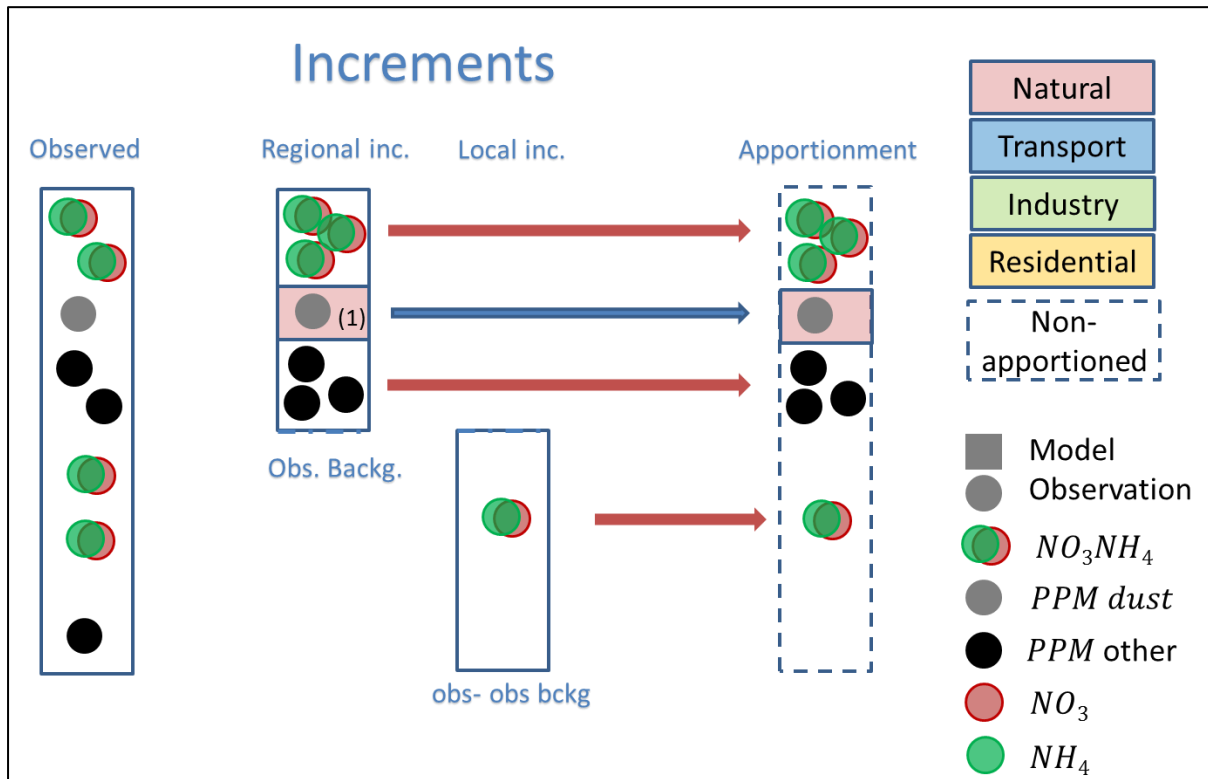


Figure 17: Process to obtain increments, starting from the measurements (left column) towards the final sectoral apportionment (right column). Sources that can be apportioned in terms of their sectoral origins are shaded with colours indicated in the top-right legend. Increments are measurement-based (circles). The background increment is equal to the measured background concentration (2<sup>nd</sup> column) while the local increment (3<sup>rd</sup> column) is obtained by difference between the observations (1<sup>st</sup> column) and the background increment. As increments are only spatial, no sectoral apportionment is provided (right column). Dust is apportioned directly from measurements as it originates from a single spatial and sectoral source.

The incremental approach is not able to provide a sectoral apportionment.

## 5.5. Comparative overview

Source apportionment sectorial components as calculated from the different SA approaches applied to our example are summarized in Table 1 and Figure 18. We focus our comparison on the residential sector (similar conclusions apply to the industrial sector).

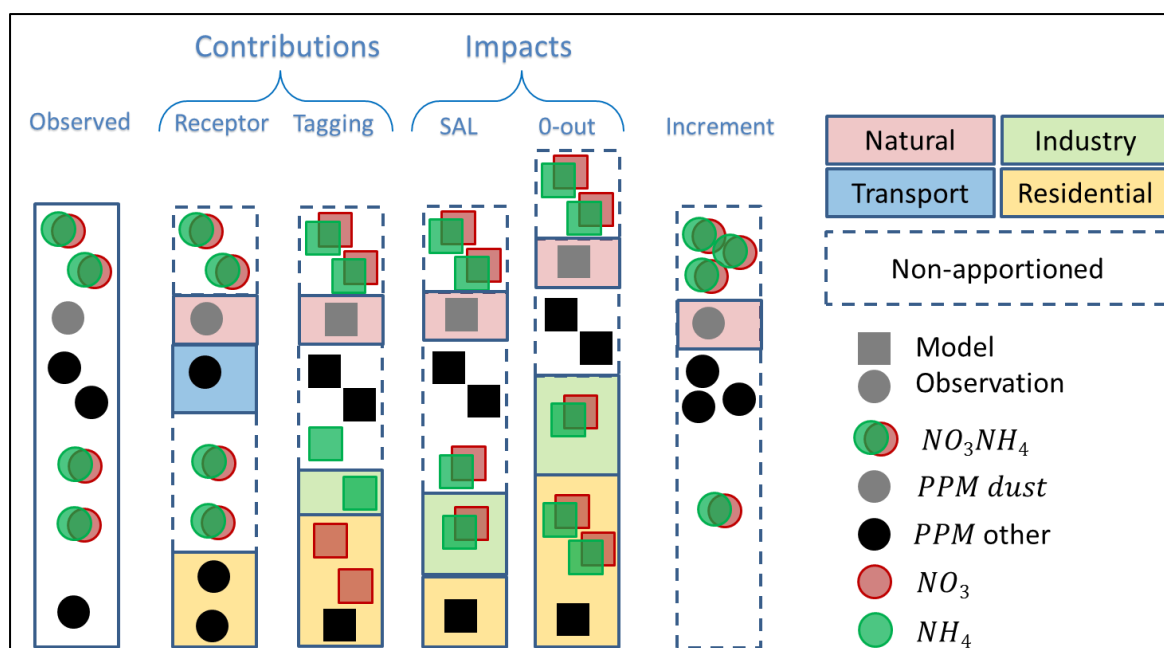


Figure 18: Graphical overview of the sectorial source apportionment components obtained with different approaches.

|                       | Residential component | Industrial component |
|-----------------------|-----------------------|----------------------|
| Receptor contribution | ● ●                   | None                 |
| Tagged contribution   | ■ ■ ■                 | ■                    |
| Zero-out Impact       | ■ ■ ■ ■               | ■ ■ ■                |
| Src. Alloc. Impact    | ■                     | ■ ■ ■                |
| Increment             |                       |                      |

Table 1: Overview of the formulations and values for the source apportionment components obtained by different approaches applied to the local industry and residential sectors. See explanations for symbols in the text.

The comparative overview highlights the following differences:

- Model vs. measurements: in terms of sectorial apportionment, only the receptor model approach is measurement-based (circle symbols). All other approaches therefore rely on input data (specific to each method) that will determine the quality of the apportionment but also the differences between the model-based approaches themselves.
- Local vs background: Although this section is about sectorial apportionment, it is important to note that model-based approach only apportion the sources that emit within a given model domain while measurement-based approaches sectorally apportion the overall (background + local) source. For example, the primary residential component is 2 (circles) for the receptor while it is 1 (squares) for other approaches. Along the same line, only receptor



models quantify the transport contribution because all other methods do not have transport emissions within the modelling domain.

- Treatment of non-linear species: The receptor approach does not manage non-linear species (no red/green symbol); tagging only considers direct effects (only red symbols) while impacts consider both direct and indirect effects (mixed red-green symbols). This has important implications on the fitness-for-purpose of SA methods.
- For non-linear species, impacts depend on the intensity of the emission reduction, as illustrated by the difference between zero-out (4<sup>th</sup> columns) and source allocation (5<sup>th</sup> columns).
- For a source that is unique in terms of origin (dust in our example), all methods, including those restricted to measurement, manage the apportionment. Some do based on measurements (receptor and increment) while others do based on models.
- The incremental approach does not deliver a sectorial apportionment.

#### **In terms of property:**

Receptor contributions are measurement-based, additive and unambiguous by construction. Because the approach is limited to linear species, dynamicity is ensured but limited to these linear species.

Tagging contributions are model-based, additive and unambiguous by construction. This unambiguity is however obtained at the expense of the neglect of indirect chemical effects. Because of this neglect of indirect effects, contributions are not dynamic and prevent therefore their use to support the design of air quality plans that involve non-linear species.

As zero-out impacts arise from emission reduction, they are dynamic by construction and because they are attached to a single source, they are unambiguous. Nevertheless, these properties are obtained at the expense of a lack of additivity. For large emission reduction values, impacts are indeed not additive, when non-linear species are involved.

Similarly, to the zero-out impacts, source allocation impacts are dynamic and unambiguous by construction. Because they are calculated from a moderate enough range of emission reductions source allocation impacts remain additive (see also Section 6 for example). One of the main issues with this type of impacts is however to identify their range of applicability, i.e. define the level of emission reduction for which additivity is preserved.

Increments do not apply to sectorial apportionment.

These properties are summarised in Table 3.

## **6. Spatial apportionment**

We apply a similar reasoning for a spatial apportionment and use the same starting point example (Figure 12) to assess how the different approaches distinguish what is emitted locally (i.e. within the domain of interest) from what is originating from the background. Only the final overview is shown below.

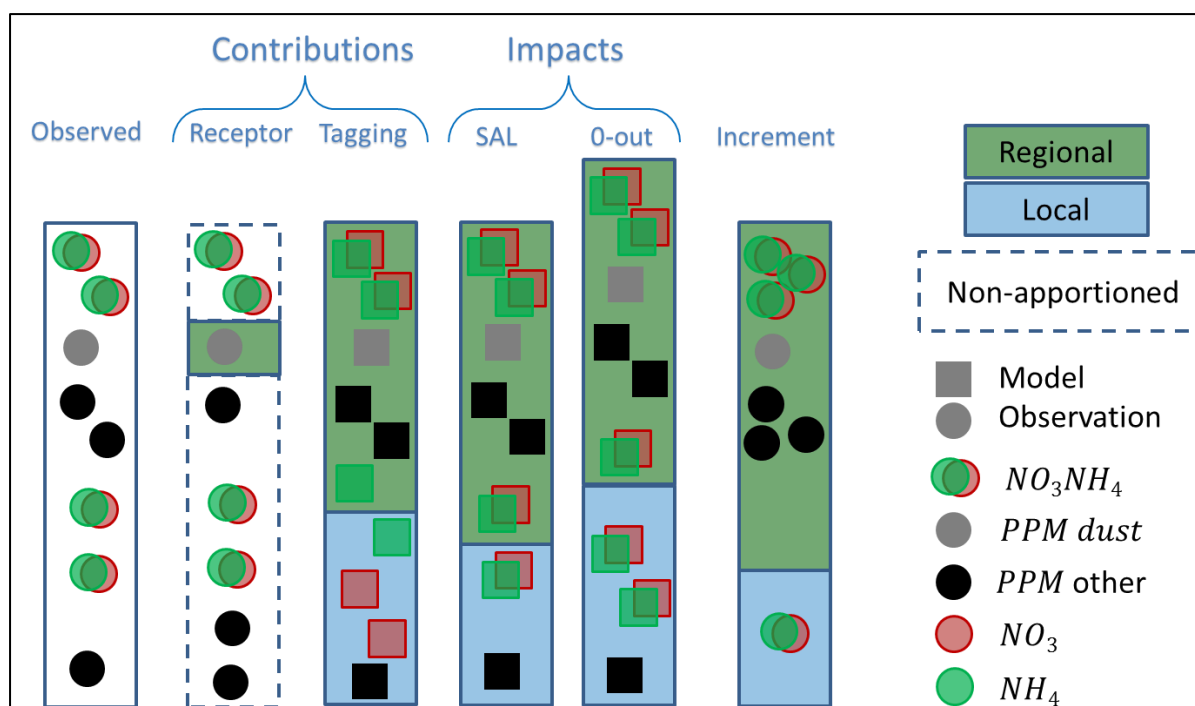


Figure 19: Graphical overview of the spatial source apportionment components obtained by different approaches (SAL = source allocation).

- By construction, receptor models (if used alone) are not able to perform a spatial apportionment and cannot therefore distinguish local from background sources contributions.
- The local tagging contribution is apportioned by tagging the local precursors (industry and residential) within the modelling domain and considering only the direct effects whenever non-linear species are involved (see previous section). Although the background contribution may be tagged, its sectorial fractions cannot be quantified.
- The local and background impacts are obtained by reducing the local emissions and the background, respectively and calculating the resulting difference on concentration. As shown by the zero-out option (4<sup>th</sup> column), one of the main issues is the lack of additivity. Indeed the sum of the local and background impacts do not sum up to the baseline concentration. Additivity is only fulfilled for limited emission reductions as for source allocation (50% reduction in our case – 5<sup>th</sup> column).
- For the incremental approach, the background component is equal to the background concentration measured away from the source. The local component is the difference between the baseline concentration and the background component, previously calculated.

The source apportionment spatial components are formulated as in Table 2.

|                              | Local component | Background component |
|------------------------------|-----------------|----------------------|
| <b>Receptor contribution</b> |                 |                      |
| <b>Tagged contribution</b>   | ■ ■ ■ ■         | ■ ■ ■ ■ ■ ■ ■ ■      |
| <b>Zero-out Impact</b>       | ■ ■ ■ ■         | ■ ■ ■ ■ ■ ■ ■ ■      |

|                    |  |  |
|--------------------|--|--|
| Src. Alloc. Impact |  |  |
| Increment          |  |  |

Table 2: Overview of the source apportionment components obtained by different approaches for spatial apportionment (based on illustrative example). See explanations for symbols in the text.

For tagging contributions and both type of impacts (source allocation and zero-impact), the properties attached to spatial SA are similar to those attached to sectorial apportionment. Receptor contributions are not applicable to spatial apportionment. On the contrary increments only apply to the spatial apportionment. They are additive by construction. However, the incremental components are unambiguously associated to the sources only when two specific assumptions are fulfilled (see Section 4.2 and Annex A). When these assumptions are not met, increments become ambiguous as they include a mix of influences from different sources. This ambiguity implies that increments are not dynamic because they do not reflect concentration changes resulting from emission changes. These properties are summarised in the Table below.

|             | Impacts  |                   | Contributions                    |         | Increment<br>(only for spatial) |
|-------------|----------|-------------------|----------------------------------|---------|---------------------------------|
|             | Zero-out | Source allocation | Receptor<br>(only for sectorial) | Tagging |                                 |
| Additivity  |          |                   |                                  |         |                                 |
| Dynamicity  |          |                   |                                  |         |                                 |
| Linearity   |          |                   |                                  |         |                                 |
| Unambiguity |          |                   |                                  |         |                                 |

Yes   
 Yes but only for linear species   
 No   
 Only valid over a limited range of reductions

Table 3: Summary of properties attached to SA approaches. Note that receptor contributions only apply to sectorial apportionments whereas increments only apply to spatial apportionments.

All source apportionment methodologies presented before are based on measurement or/and modelling data. As such, they are all affected by uncertainties (e.g. concerning the location of the measurement stations with the incremental approach or by the quality of the model and model input data). While the accuracy of the apportioned components will improve with better quality data (measurement and/or modelling), it is important to stress that the discrepancies observed between impacts, contributions and increment will remain because they are different concepts.

## Part III: Which source apportionment method for which purpose?

In this third part of the document, we analyse the fitness-for-purpose aspects of the SA methods introduced above. We distinguish mostly two purposes: (1) the use of SA to support the design of air quality plans and (2) the use of SA to increase the accuracy and robustness of air quality modelling systems. With these two purposes, this guide provides advices on how to use SA techniques in the overall context of air quality management. The use of SA to support to air quality plans is developed in Section 7 whereas the use of SA to support the quality assurance of modelling results is discussed in Section 8.

### 7. Support to the design of AQ Plans

Where concentrations are above the EU limit or target values, air quality plans (AQP) must be drawn according to the Ambient Air Quality Directives (AAQD art 1.18). To support this process, information about the origin of pollution must be provided (Annex XV), information that is reported in practice through the e-reporting scheme (IPR). The AAQD also mentions specific sources like transboundary pollution (art. 1.20), exceedances that can be attributed to natural sources (art. 20) or the winter sanding/salting of roads (art. 21) detailing direct implications they may have on air quality plans. The purpose of providing information on source apportionment in the context of the AAQD is therefore to support the design of air quality plans, i.e. identify the most effective air quality measures to implement.

Among the set of properties discussed and associated to the different source apportionment approaches, one is fundamental for this purpose: dynamicity.

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To support air quality planning, the source apportionment approach must be **dynamic**. This means that the source components must reflect the impact of emission changes on concentration changes.

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This implies that for non-linear species, only approaches that deliver impacts are suited to support air quality planning. Care must however be taken to fix their range of validity to avoid a possible lack of additivity (see section 4.3). In other words, and although it may require substantial computational resources (the number of simulations is linearly proportional to the number of sectors/regions to consider), source allocation is the recommended approach.

For linear species, both the approaches that deliver impacts and contributions (receptor and tagging) are suited to support air quality planning.

Although in agreement with several other studies (Burr and Zhang 2011a, Qiao et al. 2018, Mertens et al. 2018, Clappier et al. 2017, Grewe et al. 2010, 2012 ), these conclusions are important messages of this guide as tagging/labelling approaches are increasingly used in current applications to provide input to the preparation of air quality plans. This is the case, both for PM (Qiao et al. 2018; Guo et al. 2017; Itahashi et al. 2017; Timmermans et al. 2017; Wang et al. 2015, Hendriks et al. 2013) and for ozone (e.g. Borrego et al. 2016, Li et al. 2016, Wu et al. 2011). All these applications use *contributions*, despite their recognized limitation for air quality planning applications.

Finally, *increments* are generally not suited for both linear and non-linear species because their two additional underlying assumptions are frequently not fulfilled, resulting in ambiguous and non-dynamic components.

---

Due to non-linear processes, effective policies are not necessarily the ones tackling the most dominant emission source but those tackling the substance that is most scarce or binding in the pollution formation. This counter-intuitive result is difficult to communicate to policy makers. Neither the incremental approach nor the mass transfer approach will tell policy makers what measures are effective in reducing non-linear pollutants. Only simulation of various emission reduction scenarios will be able to support an effective policy strategy when non-linear processes are important. Of course, even that approach has limitations due the inevitable simplification in any model of chemical and meteorological processes, and weaknesses in emission and air quality data.

---

Although the overall recommendation to support planning is to use impacts-based approaches, not all sources behave non-linearly and particular conditions may apply to specific sources (mentioned above):

| Source                  | AAQD reference | Recommendation   |
|-------------------------|----------------|--|
| Transboundary pollution | Art. 1.20      | As country emissions involve both linear and non-linear species, the impact-based approach is recommended.   |
| Natural sources         | Art. 20        | A distinction must be made between linear (e.g. dust) and non-linear (e.g. NO <sub>x</sub> ) species. For non-linear, only impacts are suited while for linear, measurements, contributions or impacts would be valid approaches. Receptor modelling is however recommended given the uncertainty on the source strength in current inventories. |
| Winter sanding/salting  | Art. 21        | Being mostly composed of linear species, these sources can be apportioned from contributions or from impacts. Given the uncertainty on the source strength, receptor modelling is however recommended.   |

*Table 4: Main references in the Ambient Air Quality Directive (AAQD) articles that potentially call for the use of source apportionment.*

With respect to the e-reporting of source apportionment (data flow I – see summary Table 5), impacts-based approach is recommended given the non-linear nature of most sectors/sources, with the exception of the natural sources mentioned above. Note that the incremental and receptor approaches are not suited given their incapacity to provide sectorial and spatial information, respectively.

As explained earlier, impacts-based methods, source allocation in this context, only provide information up to a given threshold for which linearity and additivity of the responses can be ensured. Mertens et al. (2018) show that tagging contributions might provide additional information beyond that threshold. There is therefore a potential interest to use tagging contributions in complement to impacts to provide information that is more exhaustive. This point is discussed in the

“open issues” section. The table below summarizes the current set of recommendations on when to use specific methods for source apportionment to support planning, following the template proposed under the e-reporting scheme (reference e-reporting), here focused on particulate matter.

| PM         |             | Receptor contributions | Tagging/labelling Contributions | Source allocation impacts | Increments |
|------------|-------------|------------------------|---------------------------------|---------------------------|------------|
| Background | Transbound  |                        | As complement of impacts?       |                           |            |
|            | Country     |                        |                                 |                           |            |
|            | Natural     |                        |                                 |                           |            |
| Urban      | Traffic     |                        | As complement of impacts?       |                           |            |
|            | Industry    |                        |                                 |                           |            |
|            | Agriculture |                        |                                 |                           |            |
|            | Residential |                        |                                 |                           |            |
|            | Shipping    |                        |                                 |                           |            |
|            | Off-road    |                        |                                 |                           |            |
| Local      | Traffic     |                        |                                 |                           |            |
|            | Industry    |                        |                                 |                           |            |
|            | Agriculture |                        |                                 |                           |            |
|            | Residential |                        |                                 |                           |            |
|            | Shipping    |                        |                                 |                           |            |
|            | Off-road    |                        |                                 |                           |            |

Table 5: Recommendations regarding the use of SA approaches to produce e-reporting (Green=recommended; Red=non recommended; Orange=only as complementary information)

## 8. Support to the quality assurance of AQ modelling

In this section, we discuss possible comparisons between SA approaches and detail the information and benefit we can retrieve from such comparisons. We distinguish the following methods: receptor and tagging/labelling contributions, impacts and increments. For each cross comparison we detail the potential issues, provide advice on how to address them and finally discuss how this comparison can bring benefit in terms of validation or increased robustness of the air quality modelling system.

### 8.1. Tagging vs. receptor contributions

Let's start with the example of a comparison between receptor and tagging/labelling contributions (a similar reasoning applies to the comparison between receptor contributions and impacts). From the illustrative example, we know that the main points that differentiate the results obtained with the two methods are the following (*potential issues*):

1. Measurement vs. model: While receptor contributions are based on measurements, tagged contributions are model based (● vs. ■ in the illustrative example).
2. Boundary conditions: Tagged contributions are attached to the sources that lie inside the actual domain of analysis while receptor contributions do not distinguish the spatial components within a contribution (●● vs. ■ in the illustrative example).
3. Non-linear fraction: RMs do not allow to perform a source apportionment of non-linear pollutants, such as secondary inorganic aerosol (SIA) which are usually handled as aggregated “SIA” sources, i.e. not related to a specific emission category. Differently, SMs can tag both primary and secondary compounds to a corresponding emission source (nothing vs. ■■ in the illustrative example).

Advice for the comparison: To cope with the potential issues mentioned above it is necessary to ensure the following to retrieve valuable information from the inter-comparison:

- Limited background: ensure that the strengths of the local sources to assess are important with respect to the background pollution to limit the possible confusion with pollution originating as boundary conditions.
- Linear species only: Limit the comparison to linear species.

The main benefit of the comparison resides in the possible comparison of model-based results with measurements (i.e. *model validation*). Although limited to linear compounds, this comparison can constitute a very good approach to assess the modelled and measured strengths of emission sources and improve the inventory. Although the comparison is limited to linear compounds, the information obtained about a possible overestimation/underestimation of the emission strength of a certain source for linear compounds can be used to retrieve useful information on the emission strength of the same source for non-linear compounds, if we assume that the emission factors for both type of compounds are correct.

The main points discussed in this section are synthetized in Figure 20.

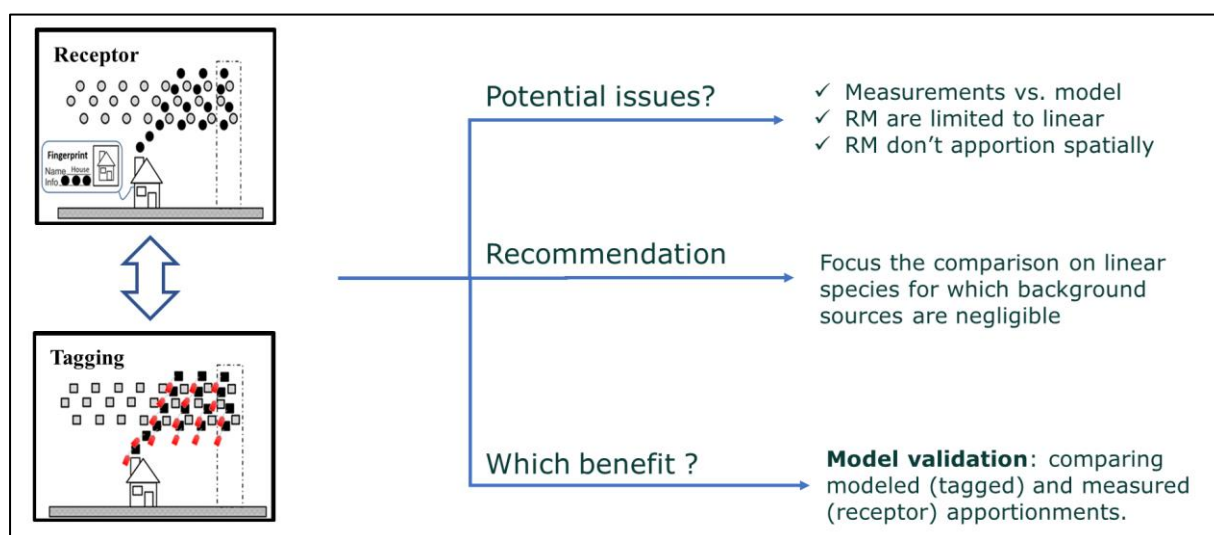


Figure 20: Synthetic overview of the potential issues (top right) met when receptor contributions (top illustration) are compared to tagging/labelling contributions (bottom illustration). The recommendation and potential benefit from the comparison are shown at the mid-right and bottom-right, respectively.

These points are also summarised in Figure 22 together with similar points drawn for the other cross-comparisons (discussed in follow-up sections). Note that some practical aspects of such comparisons are also discussed in Mircea et al. (2019).

## 8.2. Increments vs. other methods

Because increments only deliver a spatial apportionment and receptor contributions cannot, the cross-comparison of the two methods is impossible. The comparison of increments with tagging/labelling contributions or impacts is not recommended because of the following issue.

4. Increments suffer from a possible ambiguity, which importance is impossible to assess (see the two incremental assumptions described in Annex A).

## 8.3. Impacts vs. tagging/labelling contributions

For this cross-comparison, the potential issues (5 and 6 in Figure 21) consist in the following:

5. Direct/indirect effects: The main point that differentiate tagging/labelling contributions and impacts resides in the treatment of the non-linear species. As mentioned above, tagging contributions only considers direct effects, i.e. the direct links between a precursor and its product (e.g.  $\text{NO}_x \rightarrow \text{NO}_2$  and  $\text{NH}_3 \rightarrow \text{NH}_4$ ) while impacts account for indirect effects (e.g.  $\text{NO}_x \rightarrow \text{NH}_4$  or  $\text{NH}_3 \rightarrow \text{NO}_3$ ).
6. Single vs. multiple apportionments: Each impact is associated to a given emission reduction strength which for non-linear compounds will lead to different apportionment results. This is not the case for tagging/labelling contributions that represent a single apportionment.

Advice for the comparison: Existing comparisons (e.g. Grewe et al. 2012, Burr and Zhang 2011, Kranenburg et al. 2013, Clappier et al. 2017, Thunis et al. 2019) all clearly indicate that these methods deliver different results for non-linear compounds. This result is expected as the two approaches are intended to answer different questions. The advice is therefore to limit the comparison to linear species.

The main benefit of the comparison is to improve model robustness.

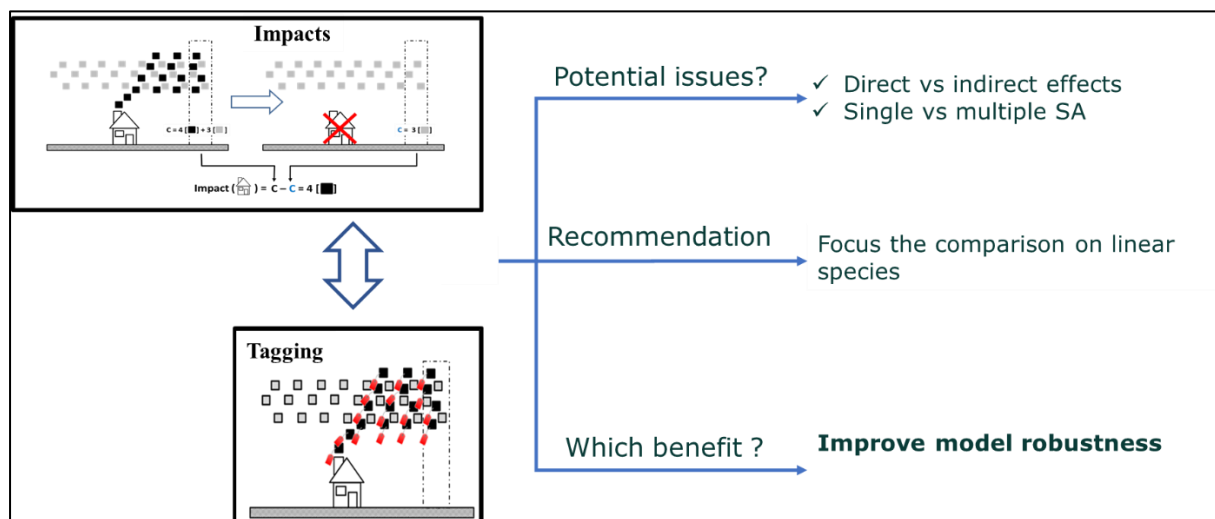


Figure 21: Synthetic overview of the potential issues (top right) met when impacts (top illustration) are compared to tagging/labelling contributions (bottom illustration). The recommendation and potential benefit from the comparison are shown at the mid-right and bottom-right, respectively



## 8.4. Comparison of methods with themselves









In general, there are no restriction on the comparison of methods with themselves (e.g. two tagging algorithm implemented in different models). The main benefit is to increase trust in the approach (model robustness). We can mention two special cases:

- Comparison of modelled vs. measured increments: Useful to assess the capability of the modelling system to reproduce spatial gradients of concentrations (ref TFMM work)
- Comparison of impacts obtained with different strength of emission reductions to assess the level of non-linearity in the model responses to emission changes.

## 8.5. Summary overview

The points mentioned in sections 8.1 to 8.4 are summarized in Figure 22.

What can we gain ?

|  | Receptor Contribution<br> | Tagging Contribution<br>   | Impact<br>                                   | Increment<br>  |
|--|--|---|--|---|
| Receptor Contribution<br> | Model robustness   | 1. Measurements vs. model<br>2. RM are limited to linear<br>3. RM don't apportion spatially<br><br>Advice: limit comparison to linear species for which background sources are negligible |  | 1. RM are limited to linear<br>2. RM don't apportion spatially<br>4. Increments to fulfill 2 assumptions prior to comparing<br>Advice: comparison is impossible |
| Tagging Contribution<br> | Model validation   | Model robustness  | 5. Direct vs. indirect effects<br>6. Impacts lead to multiple apportionments<br><br>Advice: limit comparison to linear species | 5. Direct vs. indirect effects<br>4. Increments to fulfill 2 assumptions prior to comparing<br>Advice: comparison is often misleading                           |
| Impact<br>              | comparison of modeled (tagged or impacts) and measured (receptor) apportionments.                          | Model robustness  | 6. Impacts lead to multiple apportionments<br><br>Model robustness<br>Assess non-linearities                                   | 4. Increments to fulfill 2 assumptions prior to comparing<br><br>Advice: comparison is often misleading   |
| Increment<br>           |  | Not recommended:<br><br>The two incremental assumptions are generally not fulfilled   |  | Model validation<br>compare modeled (and measured increments.   |

What should we care about?

Figure 22: The potential issues (and advice to address them) are detailed at each row/column intersection while the column/row intersection inform on the potential benefit of the comparison. The numbered bullet items refer to Section 8.1, 8.2 and 8.3. See additional explanations in the text.

## PART IV: OPEN ISSUES

### Extension to other pollutants

The guide currently focuses on particulate matter (PM) because most of the methods are usually applied for this species. Can we extend it to other species like  $\text{NO}_2$  and  $\text{O}_3$ ?

#### Questions:

- *Can we and do we have experience in using receptor models for  $\text{NO}_2$  and/or  $\text{O}_3$ ?*
- *Are there experiences using incremental methods for  $\text{NO}_2$  and/or  $\text{O}_3$ ?*
- *How can we translate our current recommendations on PM for  $\text{O}_3$  or  $\text{NO}_2$  for tagging methods?*
- *What are the linearity/additivity limits for source allocation for  $\text{NO}_2$  and/or  $\text{O}_3$ ?*
- *Can we extend the e-reporting table to  $\text{O}_3$  and or  $\text{NO}_2$*

### Distinction between linear and non-linear pollutants

The fitness-for-purpose of a SA approach is mostly determined by whether pollutants behave linearly or non-linearly. Indeed, for linear compounds, all methods with the exception of the incremental produce similar results. It is therefore important to distinguish the compounds that behave linearly from those that do not. As the limit between the two is not always a yes/no, it is also important to discuss a margin of tolerance.

Examples of linear species would potentially include passive species that remain stable with time (e.g. primary particulate matter); species that undergo ageing processes (e.g. aged marine salt (Scerri et al. 2018)) or “linear” secondary species, as some secondary organic species (Srivastava et al. 2018; Wang et al. 2018; Zhao et al. 2018). Examples of non-linear species would be species that are affected by second or higher order chemical reactions (e.g. ozone or secondary inorganic PM).

#### Questions:

- *Can we provide a list of compounds that behave linearly (for which no issue arises) and a list of compounds that behave non-linearly?*
- *Can we provide additional information on aspects that will impact the linear/non-linear boundary, e.g. the averaging time considered for the indicators*

## Distinction between source identification and apportionment

This guide is about source apportionment but some SA methods can be extremely useful to identify or to assess a source without a full source apportionment. If we define source identification, source assessment and source apportionment as follows (Figure 23):

- Source identification: Is **one** given source related to my concentrations: **Yes/No**
- Source assessment: What is the importance of **one** given source (%)
- Source apportionment: what is the relative share of **all sources**?

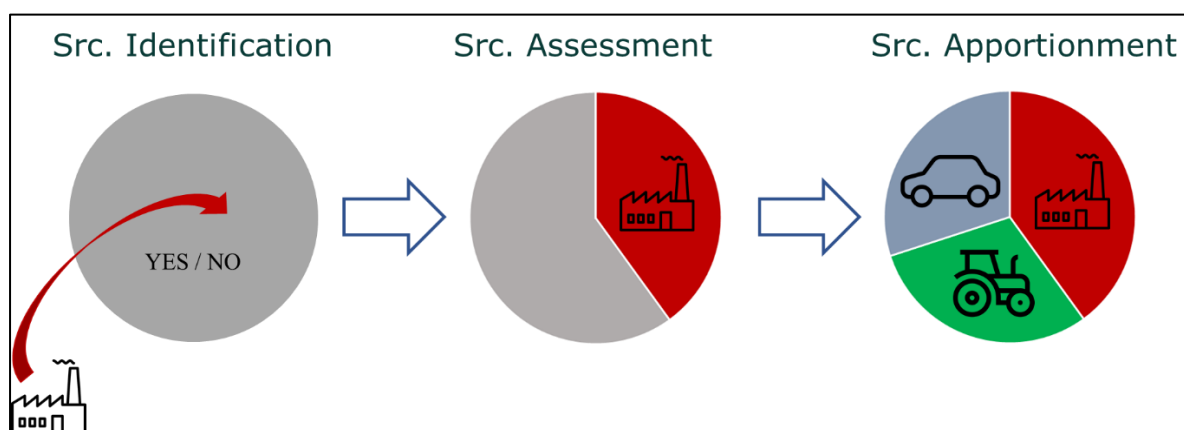


Figure 23: schematic differences between source identification, source assessment and source apportionment

As an example, receptor models are somehow capable of “apportioning” certain types of non-linear secondary compounds (e.g. classes of SOA) indirectly via other related compounds (e.g. tracers) or via their properties (analysis of the frequency response with particular instruments). This method allows to label them with respect to their origin (e.g. organic matter from fossil fuel vs biomass vs biogenic emissions). While non-linear with respect to their emission precursors, such compounds, can therefore be “apportioned” anyhow. It remains however unclear whether these advanced methods refer to source identification, assessment or apportionment, as introduced above.

### Questions:

- Although limited to linear species and to sectorial apportionments, receptor models have been very useful to identify and assess the role of sources like biomass burning, sea salt or soil resuspension that have unique chemical markers. This is especially useful to improve air quality modelling, as these sources are generally not well represented in the emission inventories. Should we develop these distinctions further?
- Can we provide information on the direct use of measurements for the source assessment of some sources that are well known in terms of spatial and sectorial origin (e.g. wind-blown dust). Can we detail for which sources this would be valid and how to proceed?

## Combined source allocation / tagging approach to support planning

In section 2.4, we referred to SA methods used in combination. We explore here the possibility of using tagging contributions to complement source allocation impacts in supporting AQ planning. Because source allocation and tagging lead to identical results for linear compounds, we focus our example on non-linear compounds. In this example (Figure 24), source allocation applies for emission reductions strengths up to 60% and provides for these levels a straightforward source

apportionment that is dynamic, additive and linear (see Section 4.6). This is however not the case beyond that emission reduction level.

For straight measures (i.e. measures that involve reducing one sector at a time), zero-out impacts might be used but this approach is demanding because all sectors need to be reduced one by one with specific AQM simulations. In addition the method is often non-additive and provides only a partial view because combined measures are excluded. An alternative is to use tagging contributions to identify the remaining sectors on which to act. Because contributions are not dynamic, they however cannot be used to retrieve quantitative information but may be used to prioritise actions. While source allocation impacts provide a top-down view on which actions (and associated reduction strength) will effectively reduce concentration, tagging contributions provide a bottom-up view on the sectors to prioritize, when emission reductions go beyond a given threshold (either for straight or combined measures). It is interesting to note that while source allocation only targets industry in our example (because  $\text{NH}_3$  is the limiting compound), tagging contributions target all three sources as the chemical regime is not anymore  $\text{NH}_3$  limited once some of the sources have been reduced by 70%. **It is important to note however that tagging contributions do not provide quantitative information on the impact of a given emission reduction and should only be used as a complement to source allocation when used to support planning.**

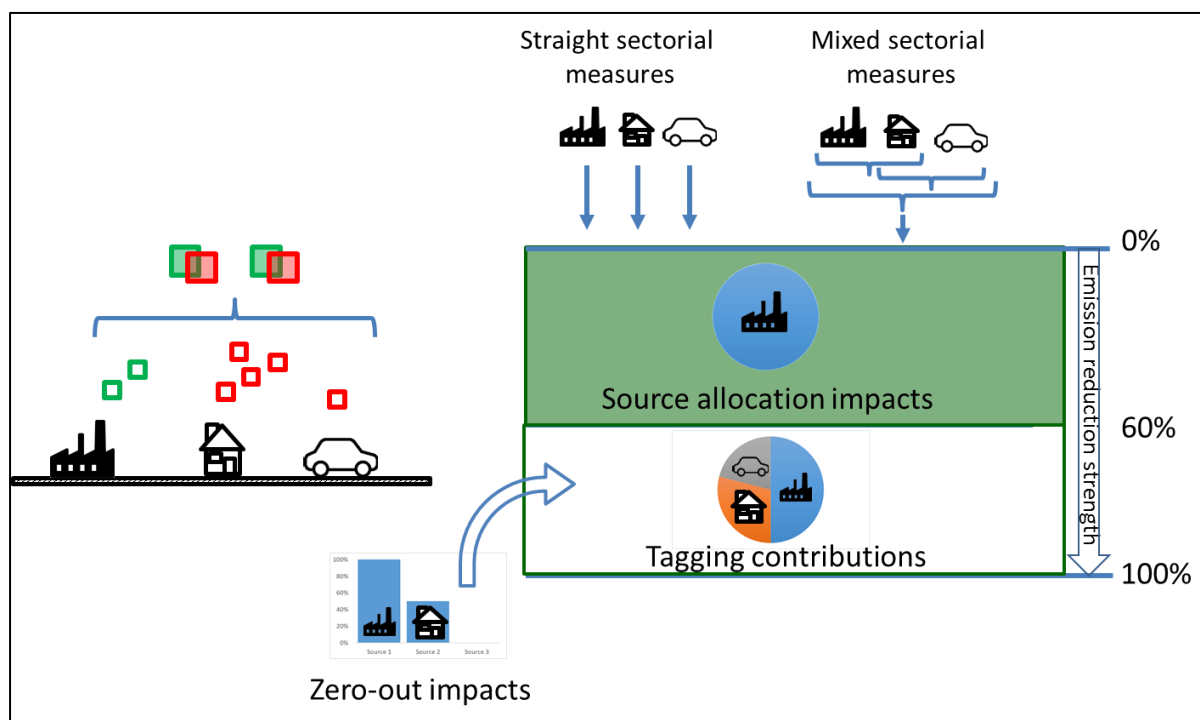


Figure 24: Three sources (left) emit either  $\text{NH}_3$  (green) or  $\text{NO}_x$  (red) compounds that combine on a 1:1 basis to form ammonium nitrate (combined solid green-red boxes). The right table differentiates straight (only one sector is reduced at a time) from mixed measures (more sectors are reduced contemporaneously). The threshold of application of the source allocation is indicated by a horizontal line (here at 60%). The green shading indicates the zones where the applied method is linear, additive and dynamic. For straight measures beyond the source allocation threshold, zero-out impacts can be calculated but are not additive (sum of contributions > 100%).

The second example (built on a similar ideas but with transport being more important - Figure 25) shows a case where zero-out impacts preserve additivity, linearity and dynamicity, providing therefore a straightforward (but resource demanding) source apportionment for straight measures. However this method cannot be used to address mixed measures beyond a given threshold for which tagging contributions provide a complementary view.

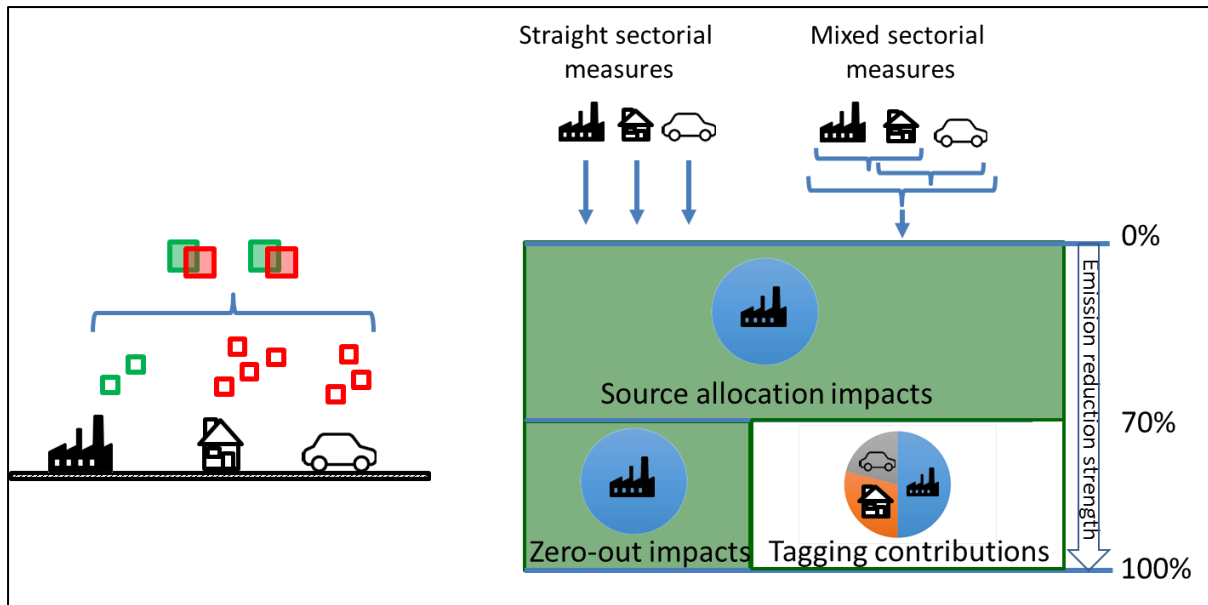


Figure 25: Three sources (left) emit either  $\text{NH}_3$  (green) or  $\text{NO}_x$  (red) compounds that combine on a 1:1 basis to form ammonium nitrate (combined solid green-red boxes). The right table differentiates straight (only one sector is reduced at a time) from mixed measures (more sectors are reduced contemporaneously). The threshold of application of the source allocation is indicated by a horizontal line (here at 70%). The green shading indicates the zones where the applied method is linear, additive and dynamic. For straight measures beyond the source allocation threshold, zero-out impacts can be calculated and is additive (sum of contributions = 100%).

#### Question:

Impact are dynamics but are very resource-demanding. On the other side, tagging methods are not dynamic but are extremely efficient from the computational point of view. Can we use tagged contribution as complementary information to the impacts to support planning? Impacts would then be used to provide dynamic information up to a threshold emission reduction while tagged contribution provide information beyond that threshold. Are the simple examples appropriate to highlight this mechanism proposed by Mertens et al. (2018)?

### Source apportionment to support the ex-post assessment of AQP

One possible purpose for SA is to assess a-posteriori whether air quality plans have been implemented or not, and if they were efficient.

#### Questions:

- Can we provide some examples and guidance on which methods to use for this particular purpose?
- How can we apply a de-trending to remove the impact of external factors like meteorology, yearly emission evolution?

## Technical Annexes

## A. Incremental assumptions

The incremental approach initially proposed by Lenschow et al. (2001) is used in many city air quality plans (Berlin2014; Segersson et al. 2017), in modelling studies (Squizzato et al. 2015; Timmermans et al. 2013; Keuken et al. 2013; Ortiz and Friedrich 2013; Pey et al. 2010) or in combined model-measurements analysis, to distinguish and quantify the street vs. the urban and/or the urban vs. the regional contributions (Kiesewetter et al. 2015). Increments (INC) are generally limited to the quantification of the spatial origins of pollution.

The urban impact (I) is defined as the change of concentration in a city due to the emissions coming from the city itself. The easiest way to express this is by imagining that all the emissions from within a city, or zone within a city are set to zero. Thus at a city location (l superscript) I is defined as:

$$I_{city}^l = C^l - B_{city}^l \quad (1)$$

where  $C^l$  is the concentration level reached at location “l” when both in-city and extra-city emissions are active and  $B^l$  is the background concentration level reached at the same location when city emissions are set to zero. The urban impact ( $I_{city}^l$ ) and the background ( $B_{city}^l$ ) explicitly depend on the size of the city over which emissions are switched off. All terms in (1) and the following relations use superscripts to indicate the location where the concentration is analysed (receptor) and subscripts to indicate the area over which emissions are switched off (source). Relation (1) can similarly be applied to any rural location (“r” superscript) at a given distance (d) from the city centre to give the impact of emissions from the city on the rural background location:

$$I_{city}^r(d) = C^r(d) - B_{city}^r(d) \quad (2)$$

The concentration at any location ( $C^r(d), C^l$ ) is then the sum of two components: an urban impact ( $I_{city}^r(d), I_{city}^l$ ) that represents the concentration due to the in-city emissions and a background level ( $B_{city}^r(d), B_{city}^l$ ) that represents the remaining concentration when in-city emissions are set to zero (Figure 1).

The urban increment and the urban impact can be connected by differencing (1) and (2):

$$I_{city}^l = \underbrace{(C^l - C^r(d))}_{\text{Lenschow increment}} + \underbrace{I_{city}^r(d)}_{\text{City spread}} + \underbrace{(B_{city}^r(d) - B_{city}^l)}_{\text{Background deviation}} \quad (1)$$

According to relation (3), the urban impact at the city centre is the sum of three components:

- The urban increment that corresponds to the concentration difference between the city centre and the rural background locations. The importance of this component depends on the distance (d) at which the rural background location is selected.
- The “city spread” that quantifies the impact of the city at the rural background location. It is equal to the urban impact at the rural background location. This component depends on distance (d) and on the size of the city fraction considered.
- The “background deviation” that quantifies any concentration difference between the city and rural background locations when the in-city emissions are set to zero. This component also depends on distance (d) and on the size of the city fraction considered....

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