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Source apportionment to support air quality management practices

A fitness-for-purpose guide (V 3.1)

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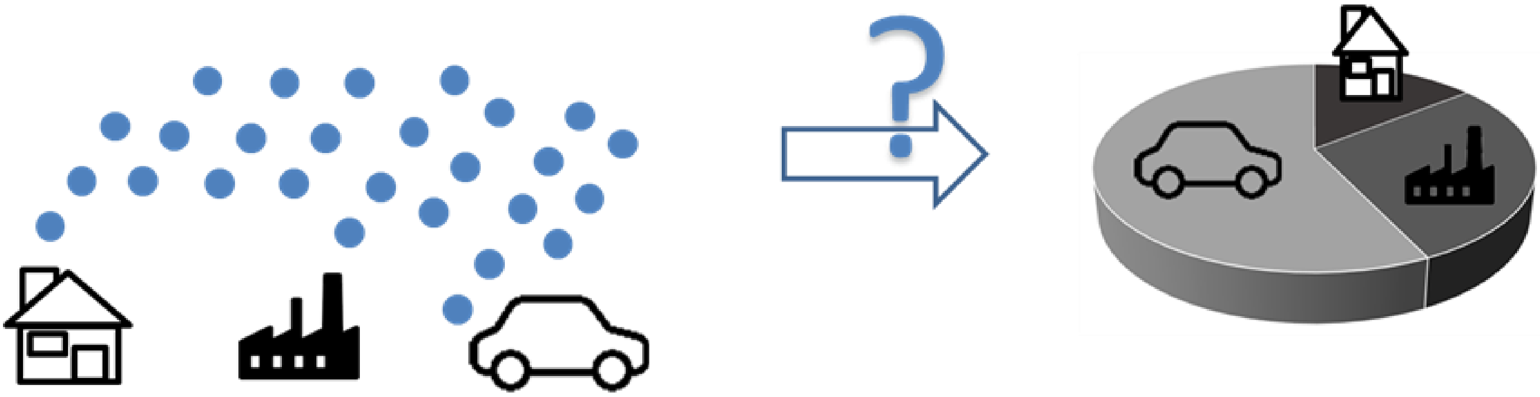

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Abstract

Information on the origin of pollution is an essential element of air quality management that helps identify measures to control air pollution. In this document, we review the most widely used source-apportionment methods for air quality management. Using simple theoretical examples we explain the differences between these methods and the circumstances where they give different results and thus possibly different conclusions for air quality management. These differences are a consequence of the assumptions that underpin each methodology and determine/limit their range of applicability. We show that ignoring these underlying assumptions is a risk for efficient/successful air quality management when the methods are used outside their scope or range of applicability. The simplest approach based on increments, contributions obtained through receptor models or tagging approaches built in air quality models as well as impacts obtained via "brute-force" methods are discussed. The guide is organised as follows: the different source apportionment methods and their associated properties are presented in Part I, simple examples are introduced in Part II to illustrate the main differences in terms of results while Part III focuses on the fitness-for-purpose aspects of the different methods. Finally, Part IV lists and briefly discusses a series of open issues.

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 (PM2.5) alone (EEA, 2018). One of the main challenges in improving this situation is to understand the origins of the pollution in order to ensure that air quality plans target the appropriate sources at the right scales to give effective results. Source apportionment is used to meet 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.

Source apportionment is a technique used to relate emissions from various pollution sources to air pollution concentrations at a given location and for a given time period

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

This document aims to support organisations in charge of air quality management in the context of the EU Ambient Air Quality Directives (AAQD). In particular, it provides information on the different source apportionment approaches that are currently in common use, describes their main characteristics and discusses their fitness-for-purpose. Finally, it also aims to support the interpretation of source apportionment results.

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

This guide is structured around four 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-purpose of the different approaches. Finally, open questions are discussed in the fourth section.

PART I: METHODS AND CONCEPTS

2 Which methods to use for source apportionment?

We distinguish three main types of source apportionment results that we refer to as: (1) **potential impacts**, (2) **contributions** and (3) **increments**. Different types of results can be used in combination. In order to highlight the differences between approaches, we use a number of simple examples in which pollutants remain stable with time, i.e. they do not undergo chemical reactions.

2.1 Potential impacts

Impacts are concentration change resulting from emission changes. They are best 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) and in Mircea et al. (2019). The method used to calculate impacts is variously referred to as "brute-force", "sensitivity analysis" or "perturbation method. The potential 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 α, divided by α, i.e.:

$$Potential\ Impact = \Delta C(\alpha)/\alpha$$

A potential impact based on α=1 is representative of a complete switch-off of the activity and is also referred to as a full impact. The division by the factor α is a mean to extrapolate virtually the impact resulting from any percentage emission reduction to 100%, hence its name "potential impact". If species are not involved in complex chemical processes (discussed in section 3) potential impacts calculated at any percentage of emission reduction are consistent (i.e. similar) implying that concentration changes are proportional to the emission reduction (i.e. a 50% emissions reduction leads to half the concentration change than a 100% emission reduction). If species are involved in complex chemical processes (discussed in Section 3), potential impacts calculated at different α values will however differ.

Potential 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 emissions reduction factor (ranging from 0 to 1).

The method to obtain potential and full impacts, that can be applied to any pollutant, is shown schematically in Figure 1 below, where square symbols indicate that impacts are model based.

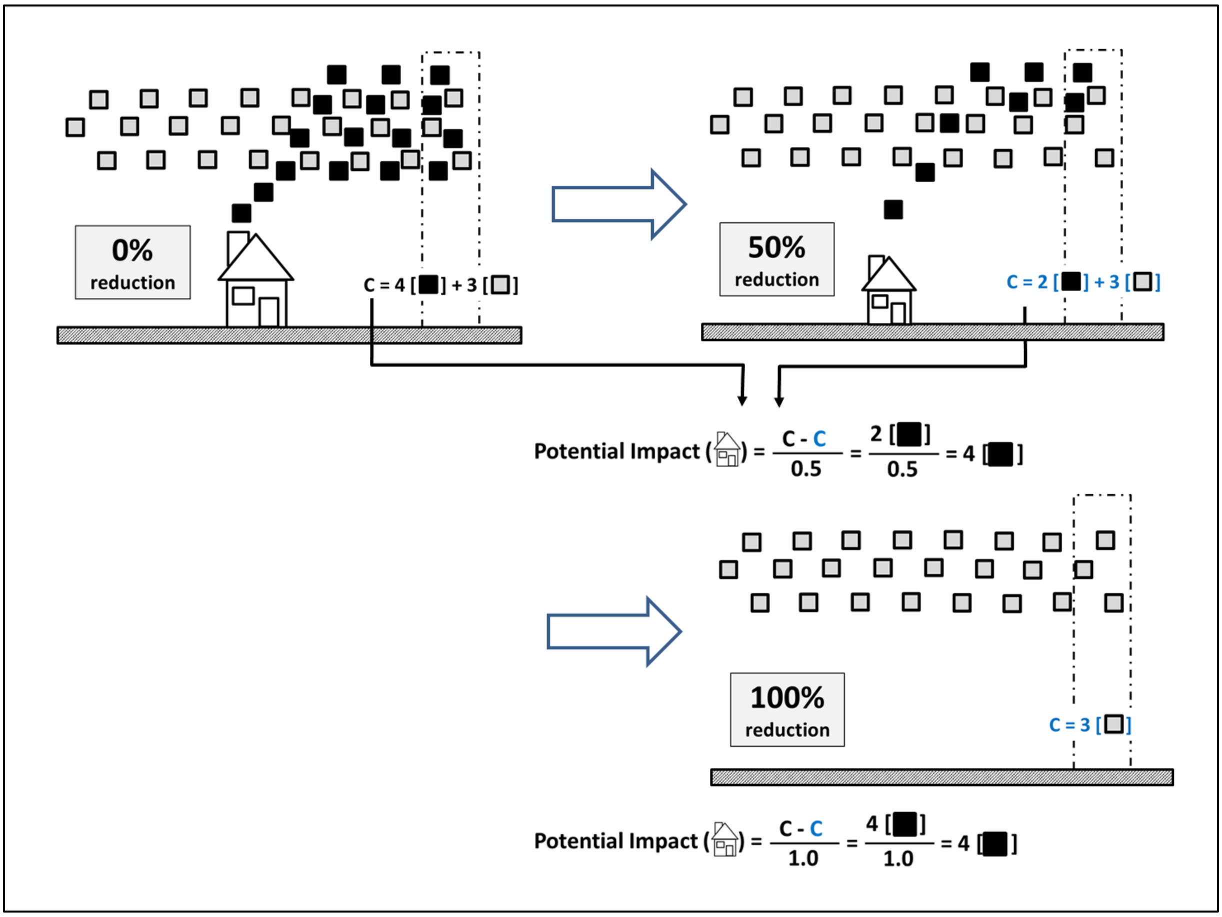

Figure 1: In this example, residential emissions (black squares) mix with the background pollution (grey squares) and lead to a given concentration downwind of the source (right dashed rectangle). When the source is reduced by 50% (right top), two out of the four black squares remain together with the background while for a full reduction, only the background remain (right bottom). Potential 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. Note that in this figure, each symbol (circle or square) represents a unit of mass that may come from the background or may be emitted by a source. With this representation, pollutant concentrations can be computed by summing up the symbols with a given volume of air at a given receptor location (e.g. the dashed lines rectangle).

2.2 Contributions

Contributions can be calculated either starting from measurements (via receptor-oriented models) or starting from model results (source-oriented models using a tagging algorithm). The differences between these methods 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 VOCs and particulate matter.

For source-oriented models (Figure 2 – right), source precursors are tagged within an AQM. For chemically reactive components, a set of reactive tracers is introduced in order to follow the evolution of source contribution through chemical pathways The labels can be defined flexibly, discriminating variously between countries/provinces, sectors or fuel types etc.. Hence, besides the concentration of each tracer, the corresponding fractional contribution of each label is also 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.

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

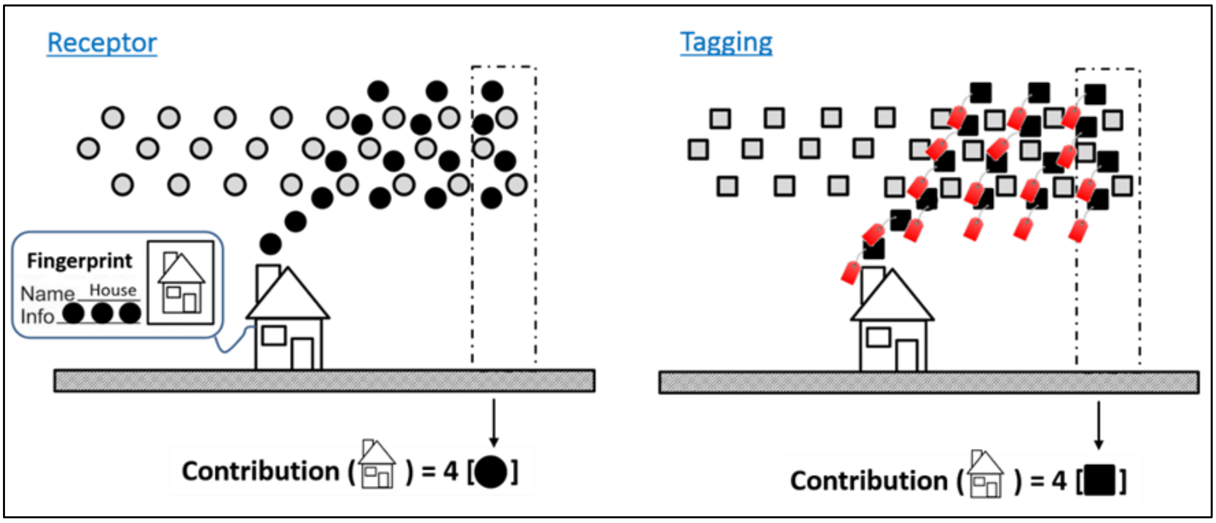

Figure 2: In this example, residential emissions (black symbols) mix with the background pollution (grey symbols) and lead to a given pollutant concentration downwind of the source (dashed rectangle). Contributions are obtained by (1 - left): recognising in the downwind concentration (via pre-established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

The incremental approach relates an emission from a source to the concentration at a given receptor by differencing the concentration at the receptor and the concentration at a nearby location that is not influenced by the source. Increments are most often calculated using measurements. The method 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.

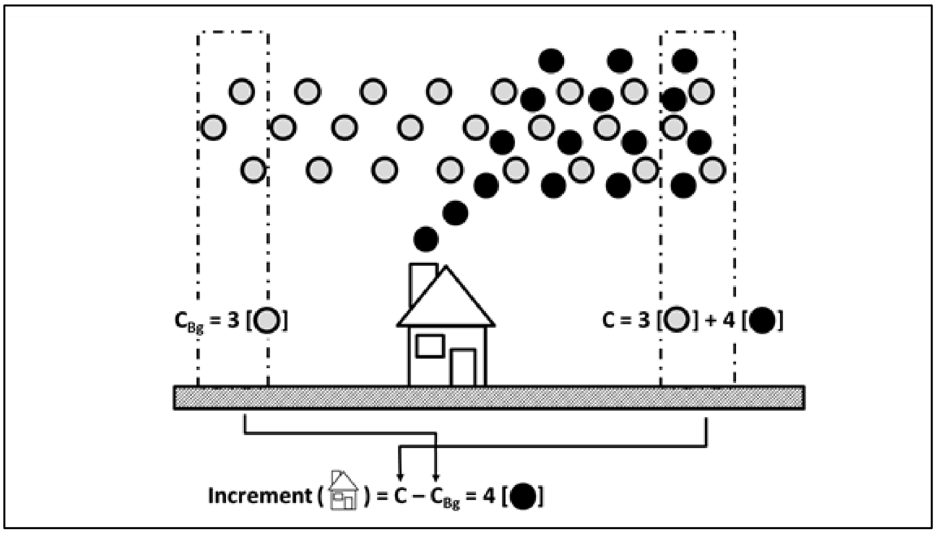

Figure 3: In this example, residential emissions (black circles) mix with the background pollution (grey circles) and lead to a given pollutant concentration downwind of the source (right dashed rectangle). Increments are obtained by subtracting the background concentration (CBg, left dashed rectangle) from the concentration C 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.

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).

Although increments are mostly measurement-based, they can also be obtained via AQMs.

2.4 Combined methods

Source apportionment applications often use a combination of methods. In designing some air quality plans, the urban and regional components are identified by increments, while potential impacts are computed in a second step to identify and quantify the sectoral origins of the pollution (Berlin 2014, Segersson et al. 2017).

Mertens et al. (2018) use potential impacts and contributions together, the former to assess the efficiency of mitigation measures on O_3 levels and the latter to retrieve additional information on unmitigated emission sources (i.e. those not covered by the potential 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 what 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 approach 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.

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

Linear behaviour: To illustrate this property, we use the example of two types of primary PM (PPM1 and PPM2) that are emitted and mix in the atmosphere (Figure 4 – middle dashed rectangle). Since they do not interact with each other, the measured PM compounds (right) correspond to the emitted ones. As shown in the scatter diagram, the concentration of PPM1 (◍) is directly proportional to the PPM1 emissions (◍) and does not depend on the PPM2 emissions (●). PPM1 behaves linearly.

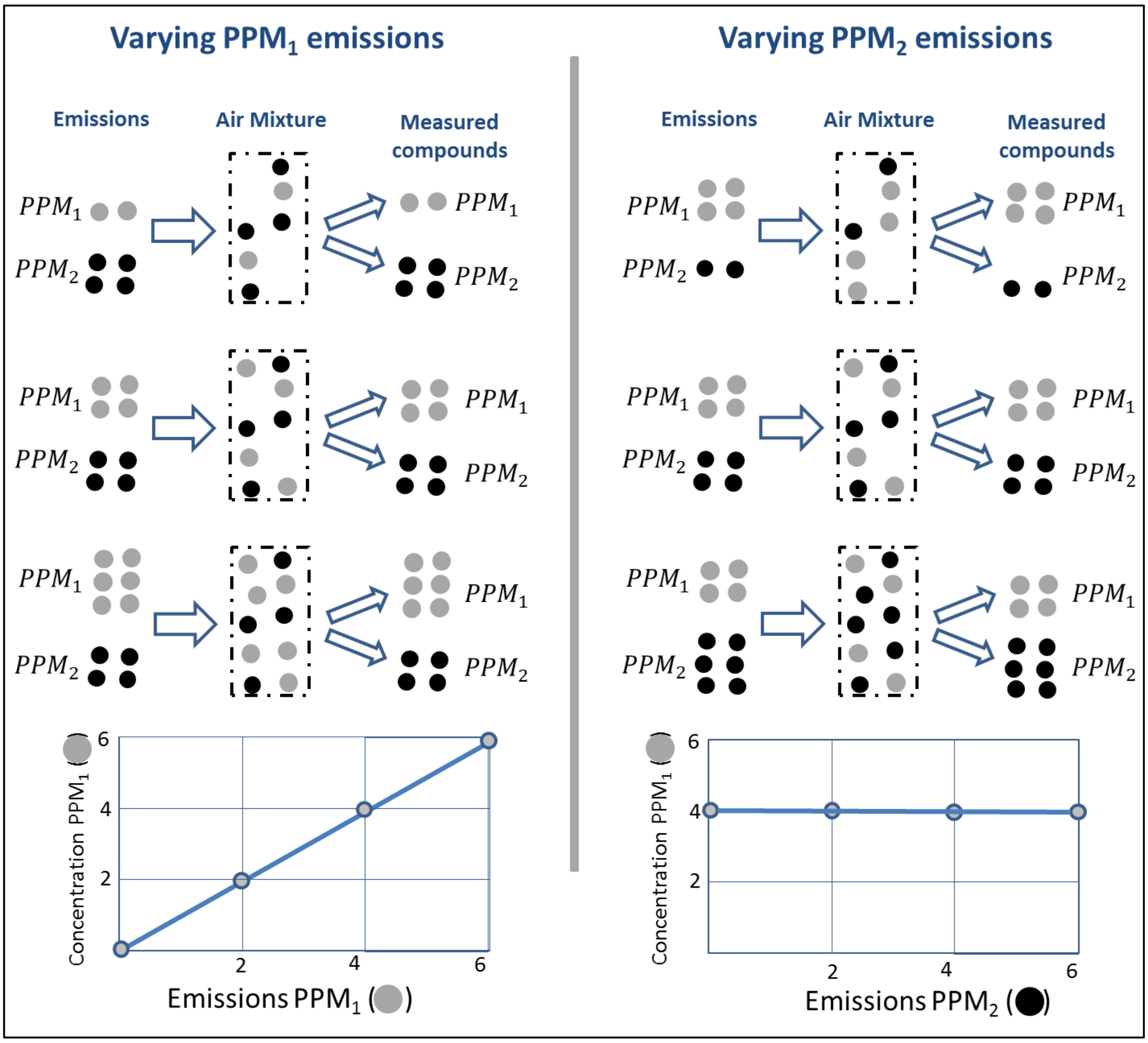

Figure 4: Schematic representation of a simple situation where two types of primary PM pollutants (PPM_1 and PPM_2) are emitted in the atmosphere and lead to specific downwind measurements. Because they do not interact with each other, the emitted pollutants are the same as those measured because the species do not interact. Situations corresponding to different shares of emissions are shown for PPM_1 (left) and PPM_2 (right).

The line plots 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 (NH$_4$NO$_3$): ○ + ⭕ → 🟢🔴. Out of ammonium nitrate (🟢🔴), nitrate (NO_3^- 🔴) and ammonium (NH_4^+ 🟢) are the measured compounds (Figure 5). If we focus on nitrate (a 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.

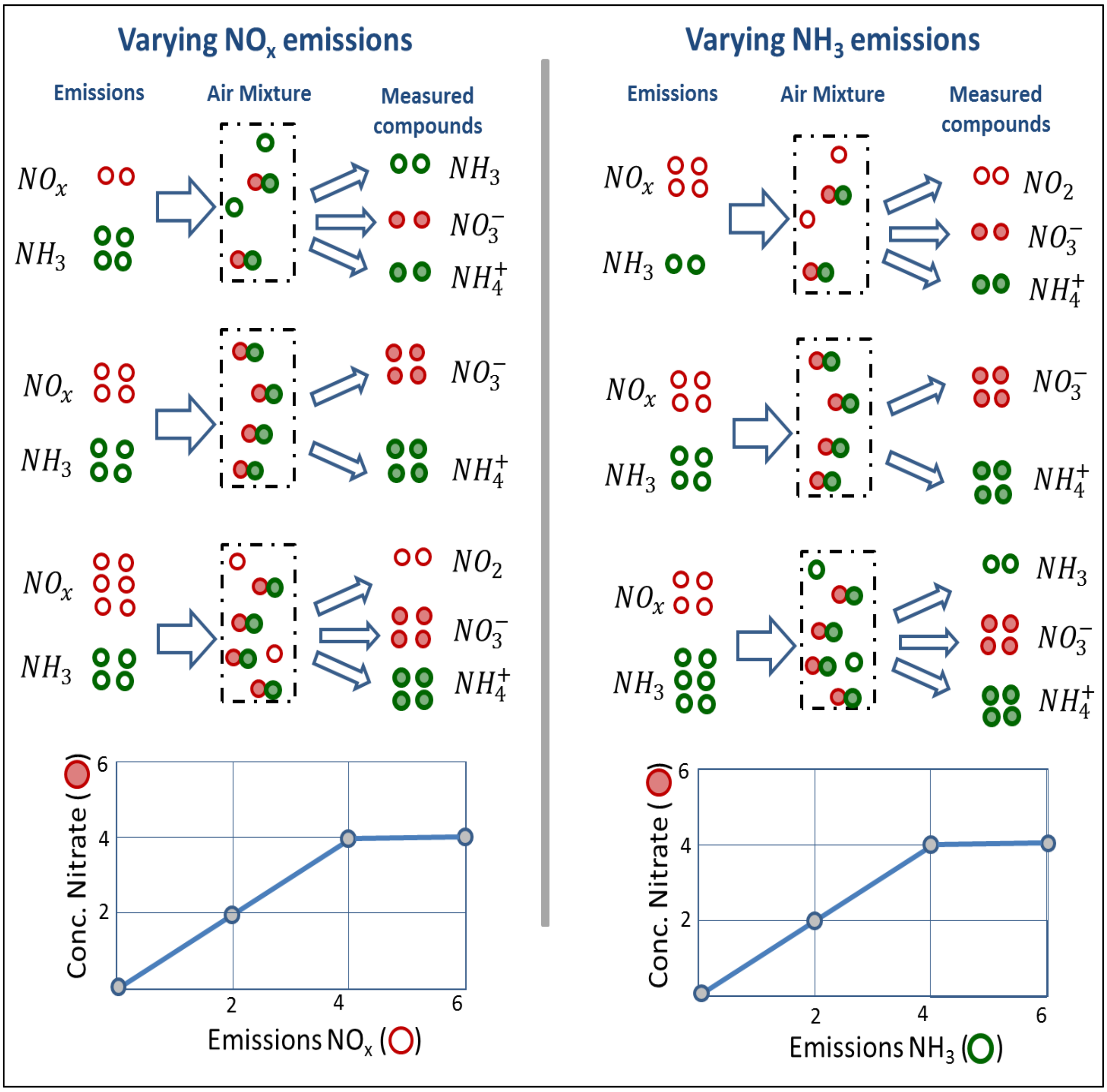

Figure 5: Simplified schematic representation of a situation where two types of gas-phase precursors: NO$_x$ (⭕) and NH$_3$ (○) are emitted in the atmosphere, react on a 1:1 basis to form ammonium nitrate (NH$_4$NO$_3$ - 🟢🔴). The atmospheric mix is then decomposed in the different measured compounds: NO$_x$, NH$_3$, nitrate (NO$_3$ - 🔴) and ammonium (NH$_4$ - 🟢). Because they interact with each other, the measured pollutants can be different from those emitted. Situations corresponding to different shares of emissions are shown for NO$_x$ (left) and NH$_3$ (right). The two scatter diagrams illustrate how the nitrate concentration ($\boldsymbol{NO_3^-}$) varies with the strength of the emissions of NO$_x$ (left) and NH$_3$ (right). Nitrate varies non-linearly in terms of both the NO$_x$ and NH$_3$ emissions. A similar behaviour (not shown) would be obtained for ammonium ($\boldsymbol{NH_4^+}$).

Indirect / direct effects:

In the example presented above, a secondary product (e.g. NO_3-) results from the interactions of different precursor emissions (NO_x and NH_3 in our case). This interaction reflects the co-dependence of nitrate (NO_3-) on both NO_x and NH_3 emissions. This is often referred as direct and indirect effects (see box below) and it is challenging for source apportionment to determine which fraction of the nitrate should be apportioned to the NO_x emissions and which fraction should be apportioned to the NH_3 emissions.

Direct / indirect effects: In the literature, the dependence of a PM chemical species (e.g. NO_3^-) to its direct precursor (NO_2) is often referred to as a direct effect while its dependence to other precursors (e.g. NH_3) is referred to as an indirect effect.

In tagging/labelling approaches this issue is solved by neglecting the indirect effects ($NO_3^- = f(NO_2)$ and $NH_4^+ = f(NH_3)$). In other words, ● is attached to ○ and ● is only attached to ○ (see application in Section 5.2).

"Local" linearity:

It is important to note that a non-linear compound can be involved in both linear and non-linear processes, depending on the range of emission strengths considered. In our example (Figure 5), nitrate responds linearly to NOx emissions in the range [0, 4] but non-linearly if the range [3, 5] is selected. This linearity over a limited (or local) range of emission reductions is used by some potential impact-based approaches (see Section 4.6).

The results of different source-apportionment methods vary when we apply them to linear or non-linear chemical species due to their intrinsic assumptions; therefore, we distinguish linear from non-linear species in our analysis. While for some compounds, it is straightforward to assess their linear or non-linear behaviour, this is not always the case and this is discussed in the "open issues" section.

4 What are the properties of individual source apportionment methods?

Before proceeding with more complex examples, we first discuss a few concepts that support our analysis, in particular the properties that we seek from source apportionment approaches. These properties will then be helpful for discussing their fitness-for-purpose. We will continue to use circles and squares to differentiate measurement-based from model-based approaches, to stress the fact that different uncertainties are associated with models, compared to measurements.

4.1 Measurement vs. Model-based

Source apportionment methods can be either measurement or model-based, or both. While incremental and receptor approaches are mostly based on measurements, potential impacts and tagging contributions can only be obtained from models.

4.2 Unambiguity

Unambiguity: A source apportionment approach is unambiguous when each component (of the concentration) 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), neither 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 increment is unambiguously related to the source in the first situation, this is not the case in the second one which has a mix of precursors from both sources (● and ○).

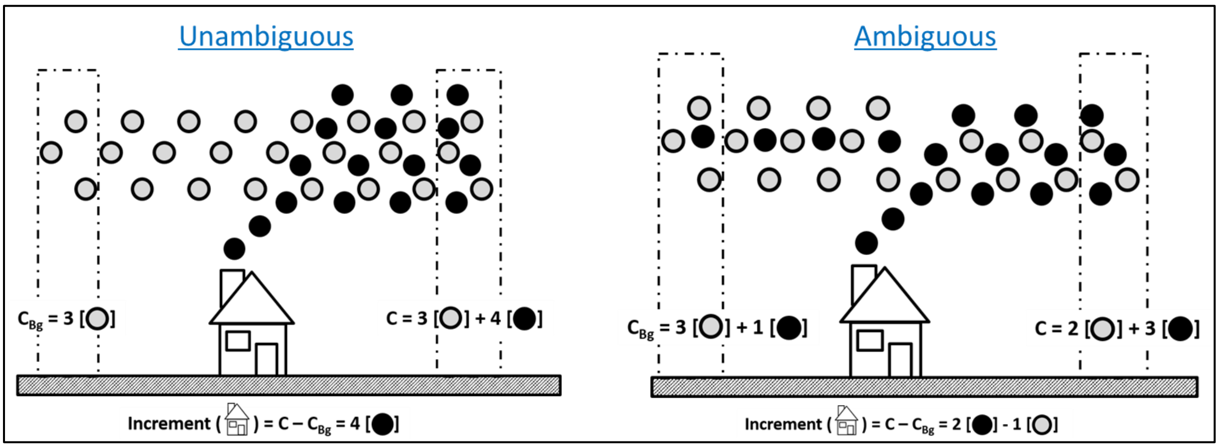

Figure 6: *Incremental approach applied to two different situations (See text for details).*

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 the Annex.

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 a potential impact-based approach would remove the ambiguity for the simple examples considered here.

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 full impact approach to the same situation as previously analysed with the addition of a constant background (Figure 7). The residential full impact on the chemical species (◧) is obtained by switching off the residential emissions (top right) and differencing with the base case. The industrial full impact is obtained similarly by switching off the industrial emissions while the combined full impact is obtained by switching off both sources contemporaneously.

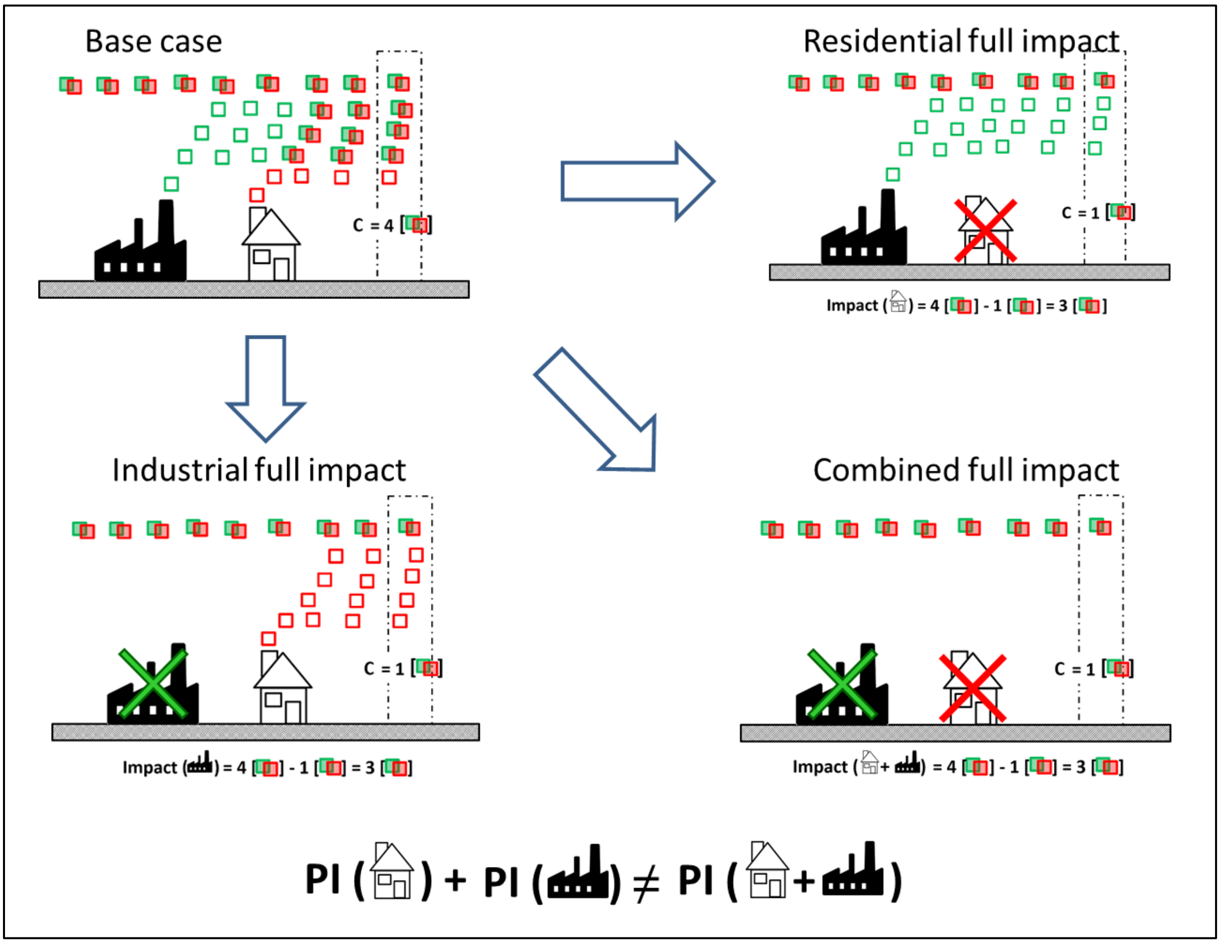

Figure 7: Assessment of the additivity. From the base case situation (top left), the industrial full impact (potential impact at α=1, denoted as PI) is calculated by subtracting from the base case the concentration obtained when industry is switched off (bottom left) while the residential full impact is obtained similarly with the residential emissions off (top right). The combined full 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 the interaction term in the literature (Stein and Alpert, 1993) because it represents the interaction between sources. As this term cannot be divided into two distinct components that relate each to one and only one source, it is *also* ambiguous.

4.4 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 a comparison between tagging contributions and potential impacts (Figure 8).

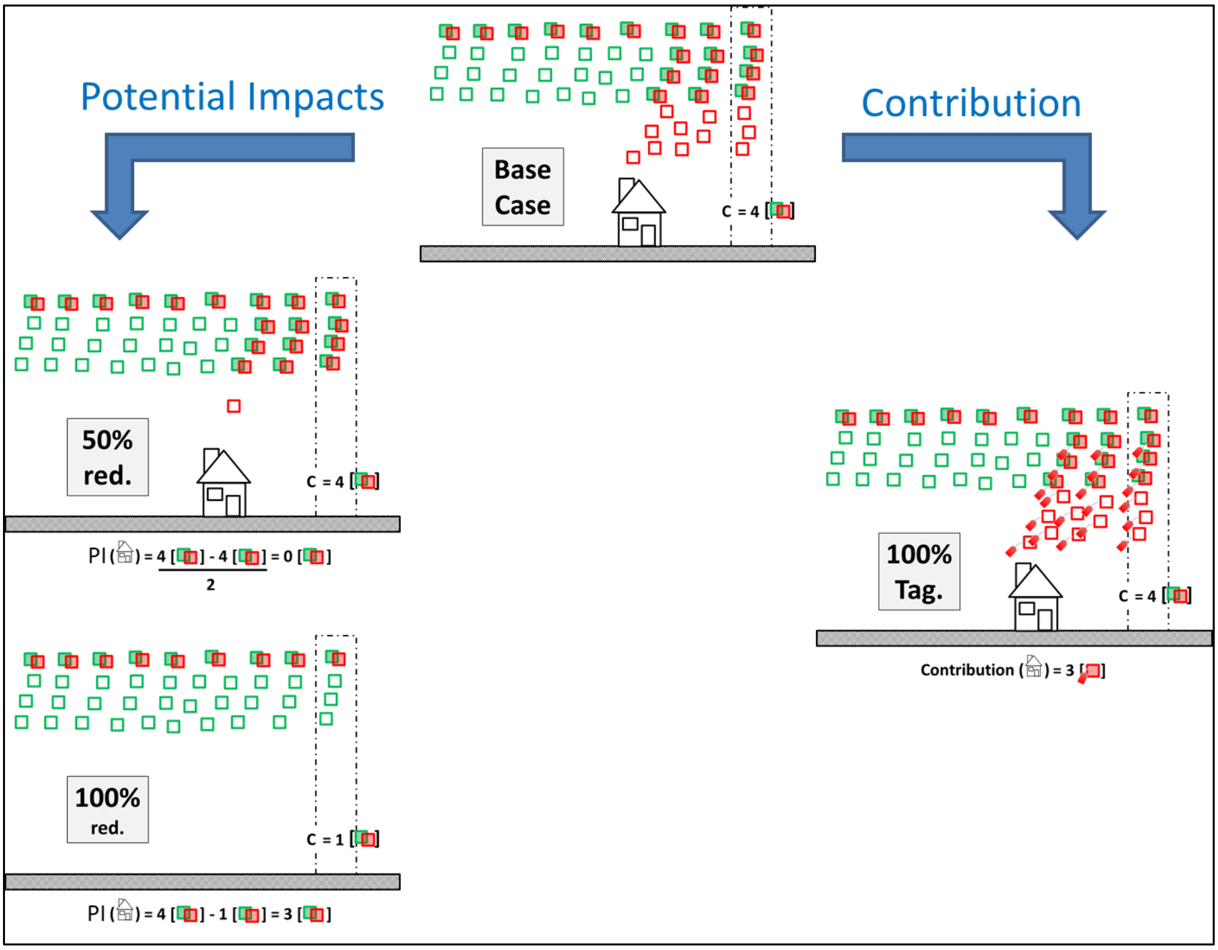

Figure 8: Example of dynamic and non-dynamic SA components. One source (residential) emits gas-phase precursors (red empty squares - NO_x) which combine with NH_3 (green empty squares) to form particulate matter (mixed shaded squares) measured at a given location (dashed rectangle). On the left side, the strength of the residential source is reduced by 50% and 100% to obtain potential impacts (PI) on PM concentrations. On the right side, the residential source emissions are tagged to obtain its contribution. In this figure, the shaded green square represents the modelled ammonium, the shaded red square the modelled nitrate and the mixed green-red shaded pair the modelled ammonium nitrate.

The references in this case are the potential impacts, because they are directly related to the impacts that reflect the consequence of emission reductions on concentration. As seen from this example, in term of mass, the tagging contribution (3 🟥) is not the same as the full impact (3 🟩🟥) because the molar masses of NO_3^- (🟥) and NH_4NO_3 (🟩🟥) are different. The tagging contribution is not the same as the 50% potential impact either. In this particular case, the 50% potential impact is zero because NO_x are not the binding compounds in the formation process (a 50% reduction of the NOx emission leaves enough NO_x molecules to react with the available NH_3 molecules). The tagging contributions are therefore not dynamic while the potential impacts are.

4.5 Consequences for 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). When the source apportionment method is additive and consistent and the species behave linearly, the pie chart is extremely easy to use to estimate the concentration change resulting from an emission reduction. The percentage of emission reduction can be simply multiplied by the percentage share of the source to be reduced to deduce this concentration change. For example, the concentration change resulting from a 50% reduction of source 2 will be equal to 50%×30%=15%. In such an ideal case, this would also apply to receptor and tagging contributions.

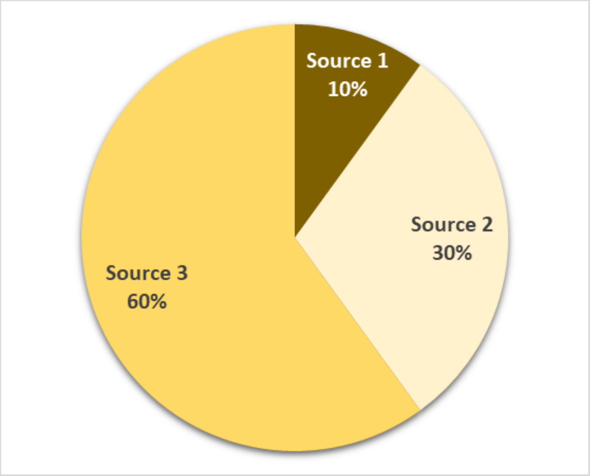

Figure 9: Example of a pie chart showing the percentage share of three sources to the total concentration

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 fulfilled, potential impacts associated to different sources cannot be summed. For example, the full impact of switching off both sources 1 & 2 will not equal the sum of the individual full impacts as indicated in the pie chart (i.e. impact (1&2) ≠ 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 becomes impossible to represent the SA results via a pie chart.
- Lack of dynamicity: If the SA approach is not dynamic, pie chart values (contributions or increments) cannot be used to determine 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 potential impact-based approaches, under certain conditions (see next Section).

- Lack of consistency: it is not possible to extrapolate the pie chart values to source strengths other 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%.

4.6 Consistent and additive potential impacts: the case of source allocation

The potential impact of a source can be estimated by switching off emissions entirely (α =100%) for a given sector/area (Osada et al. 2009; Wang et al. 2014; Wang et al. 2015; Huang et al. 2018;) or estimated by reducing emissions by a smaller amount and scaling the concentration change to 100% (e.g. multiply by five the concentration change resulting from a 20% emission reduction, assuming linear behaviour). The latter 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) for which potential impacts are consistent (i.e. have similar values) and additive are referred to as source allocation. This is explained in Figure 10 and Figure 11. Source allocation makes use of the fact that non-linear compounds may behave linearly over a limited range of emission changes, as discussed in Section 4.6.

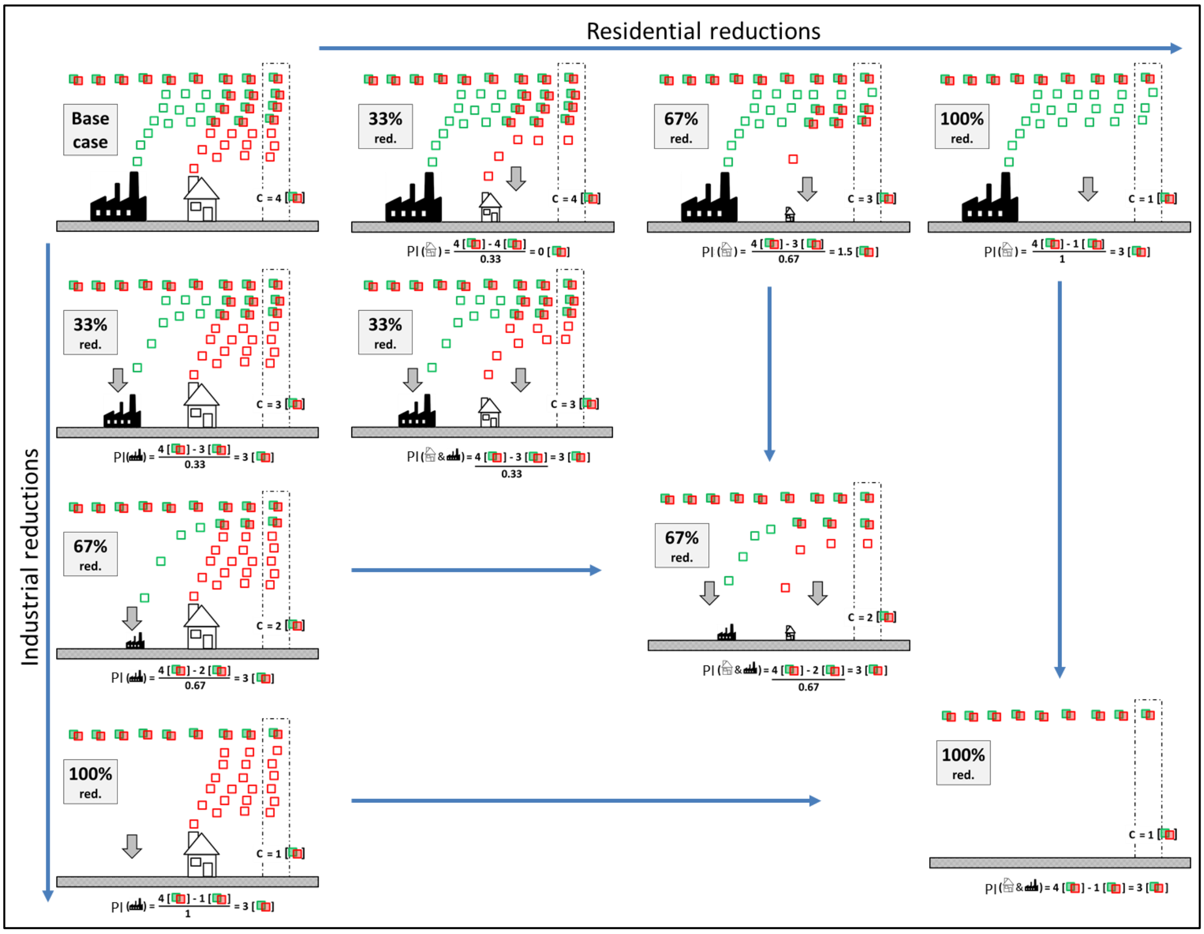

Figure 10: Potential impacts (PI) obtained for different emission reduction levels (33, 67 and 100%). The impacts are obtained by differencing the base case results (top left figure) and each of the emission reduction scenarios: the scenarios include reduction of the residential sources only (top row), 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 consistency and additivity (Figure 11).

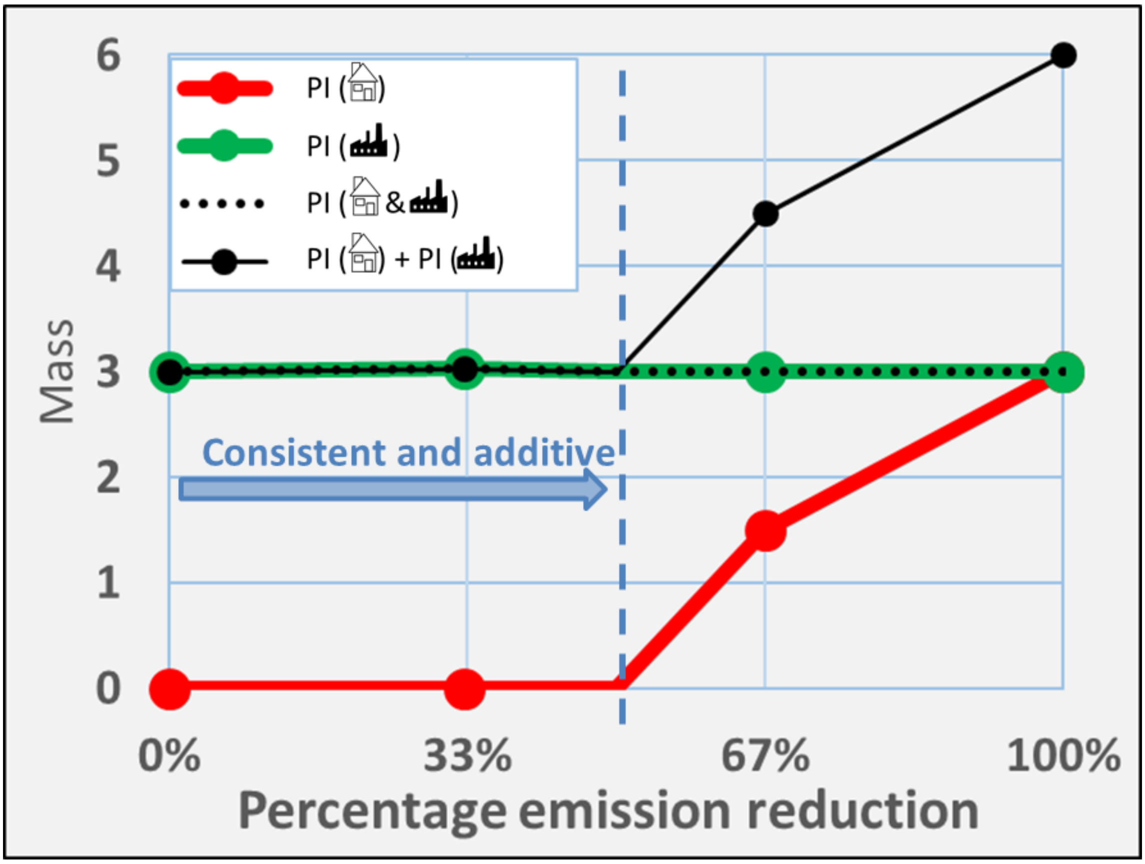

Figure 11: Based on the results of Figure 10, this diagram indicates how the residential (red) and industrial (green) potential impacts (PI) compare for different emission reduction values (X-axis). The dashed and solid lines represent the combined potential impact (both sources reduced simultaneously) and the sum of the two single potential impacts, respectively. The blue dashed line shows the application limit of the source allocation approach (left side of the diagram) that preserves consistency 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 theoretical example (Figure 12) consists of two sources (industry and residential) emitting three different pollutants, PPM (black solid circles), NO_x (red empty circles) and NH_3 (green empty circles) into a constant background composed of secondary ammonium nitrate (combined green-red shaded circles), PPM and NH_3. The background PPM originates from different sources (residential, transport and dust). As dust widely differs from the other PPM emitted by transport or residential, we represent it with another colour (grey). While PPM remains passive in this example, one mole of NH_3 reacts with one mole of NOx to produce ammonium nitrate (NH_4NO_3), a combination of nitrate NO_3^- and ammonium NH_4^+. We only focus in the following subsections on particulate matter concentrations (i.e. the shaded symbols). Note that the observations developed for each SA method refer to their stand-alone use and some of the limitations identified here could be resolved by applying SA methods in combination (e.g. receptor models with wind regression modelling; potential impacts and tagging). This particular point is addressed in Part IV.

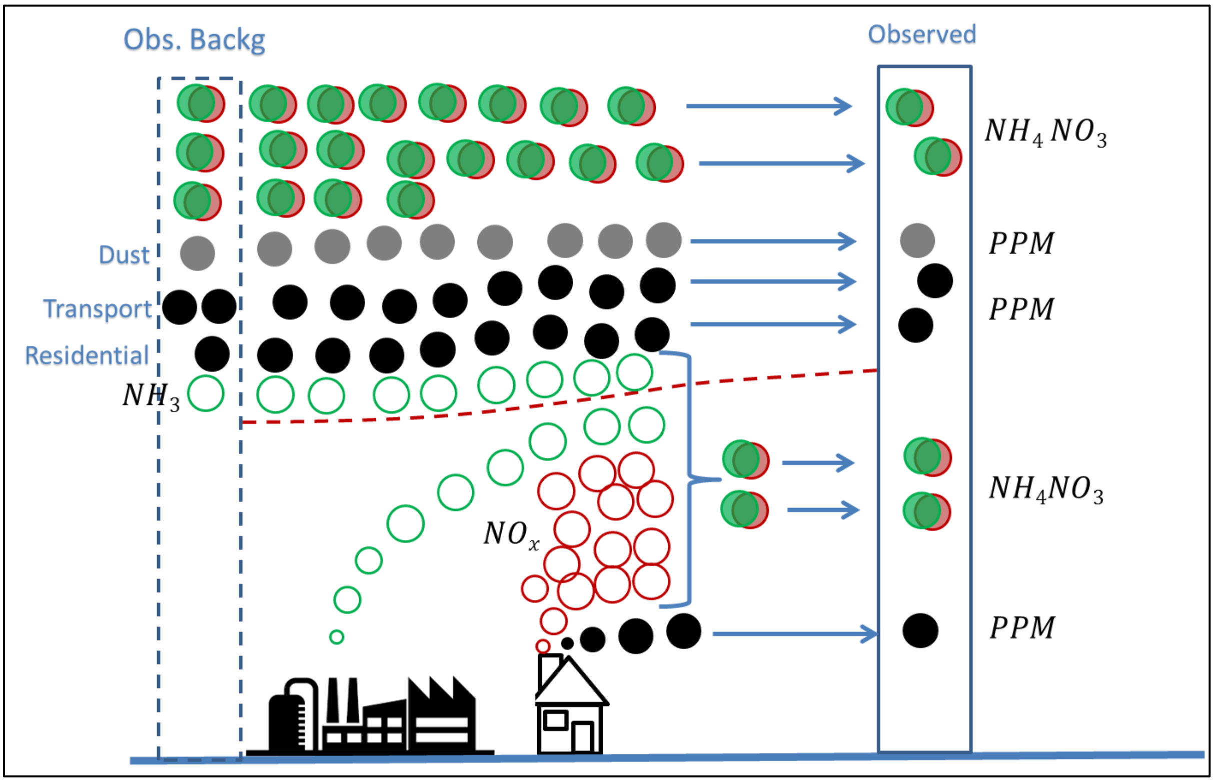

Figure 12: Illustrative theoretical example used to highlight differences between source-apportionment methods. The background pollution is distinguished from the local one by the red dashed line. The gas-phase compounds are not considered in the calculation of the final concentrations. See additional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

- Receptor models apportion the mass of an atmospheric pollutant based on measurements. We highlight this important point by keeping circle symbols for each contribution.
- 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 prior to the application of the receptor model (pink shading - point 1 in Figure 13).

- 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 only reported as concentrations and are therefore not apportioned to a well identified source (red arrows leading to dashed contoured 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)).
- In case of sources that do not modify their characteristics in space and time, receptor contributions do not differentiate between background and local (arrows 2 & 3), the residential contribution represents therefore a mix between background and local origins.

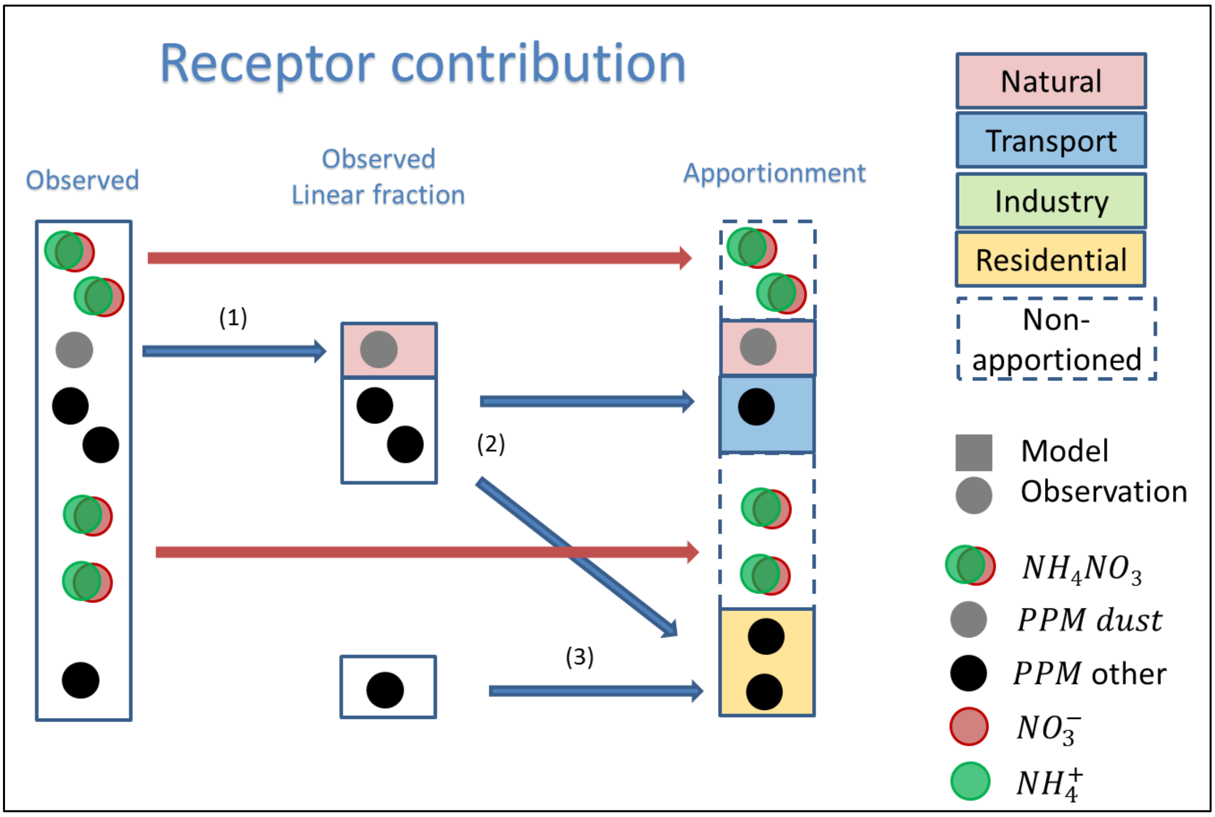

Figure 13: Process to obtain the receptor contributions, starting from the measurements (left column) towards the final sectoral contributions (right column). Sources that can be apportioned in terms of their sectoral origins are shaded with colours indicated in the top-right legend. Receptor contributions are measurement-based (circles) and only apportion linear species (middle column) and non-linear compounds are not apportioned (red arrows leading to dashed contoured 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 (1).

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. The use of different symbols highlights the fact that because of their uncertainty (or error), measurements and modelling results will differ.
- For 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 2 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 (e.g. transport) are therefore not apportioned sectorally (red arrows leading to dashed contoured rectangles). As the local sources are tagged, they can be distinguished from the background.
- For secondary products (NH_4NO_3 in our example) that result 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 (NO_3^-) in the final secondary pollutant is therefore attributed to residential activity because only this sector emits NO_x while NH_4^+ is attributed to both the industry and background.

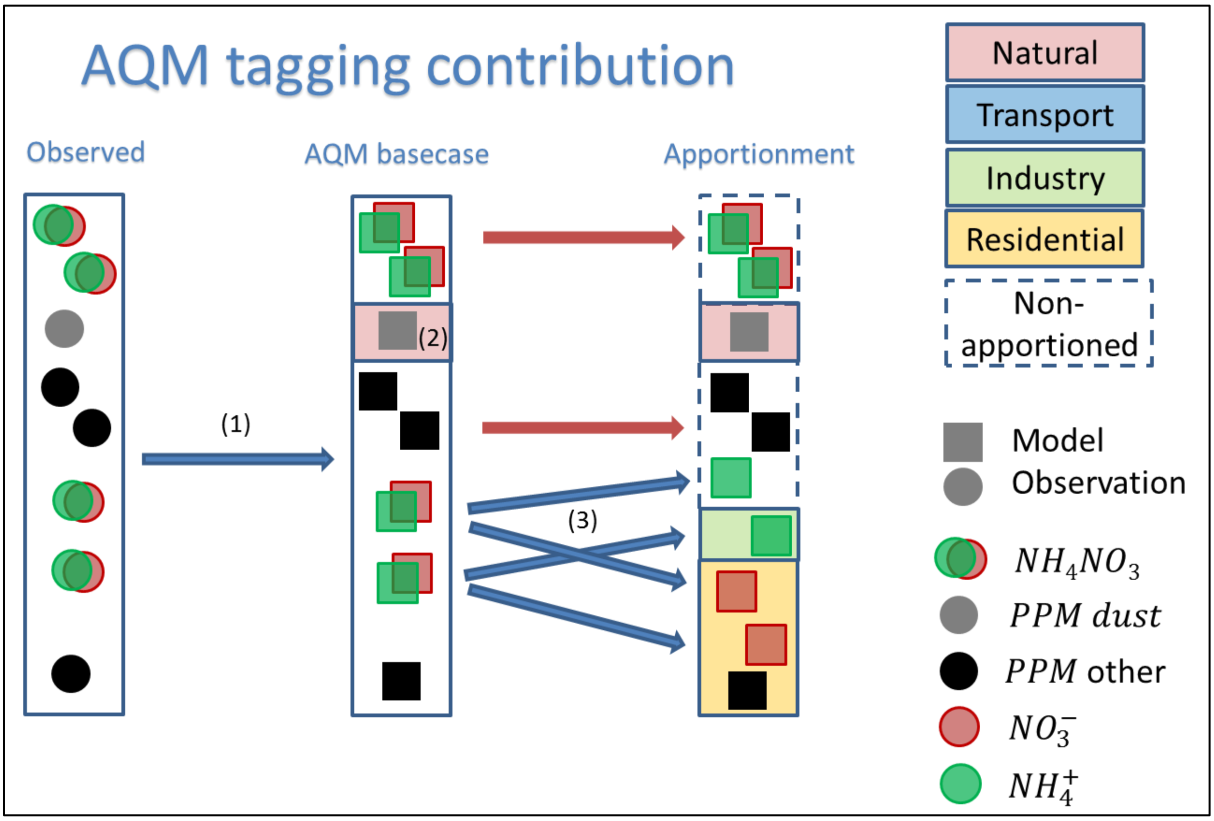

Figure 14: Process to obtain the tagging contributions, starting from the measurements (left column) towards the final sectoral apportionment (right column). Sources that can be apportioned in terms of their sectoral origins are shaded with colours indicated in the top-right legend. Tagging contributions are model-based (arrow 1 towards 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 (2).

5.3 Potential impacts

5.3.1 Full

From the observed concentration (left column in Figure 15), full impacts (potential impacts at $\alpha=1$) are obtained according to the following rules:

- Full impacts are model-based (square symbols, 2nd 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 - point 1 in Figure).
- The local industry and residential full 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 (NH_4NO_3), full impacts include an indirect effect which means that a reduction of the NO_x emissions has an influence on total ammonium nitrate, i.e. not only NO_3^- as for contributions but also NH_4^+ (arrow (3))
- Given the large emission reductions applied, additivity is not fulfilled and the sum of the full impacts and non-apportioned fractions exceed the AQM concentration (5 vs. 4 moles of ammonium nitrate).

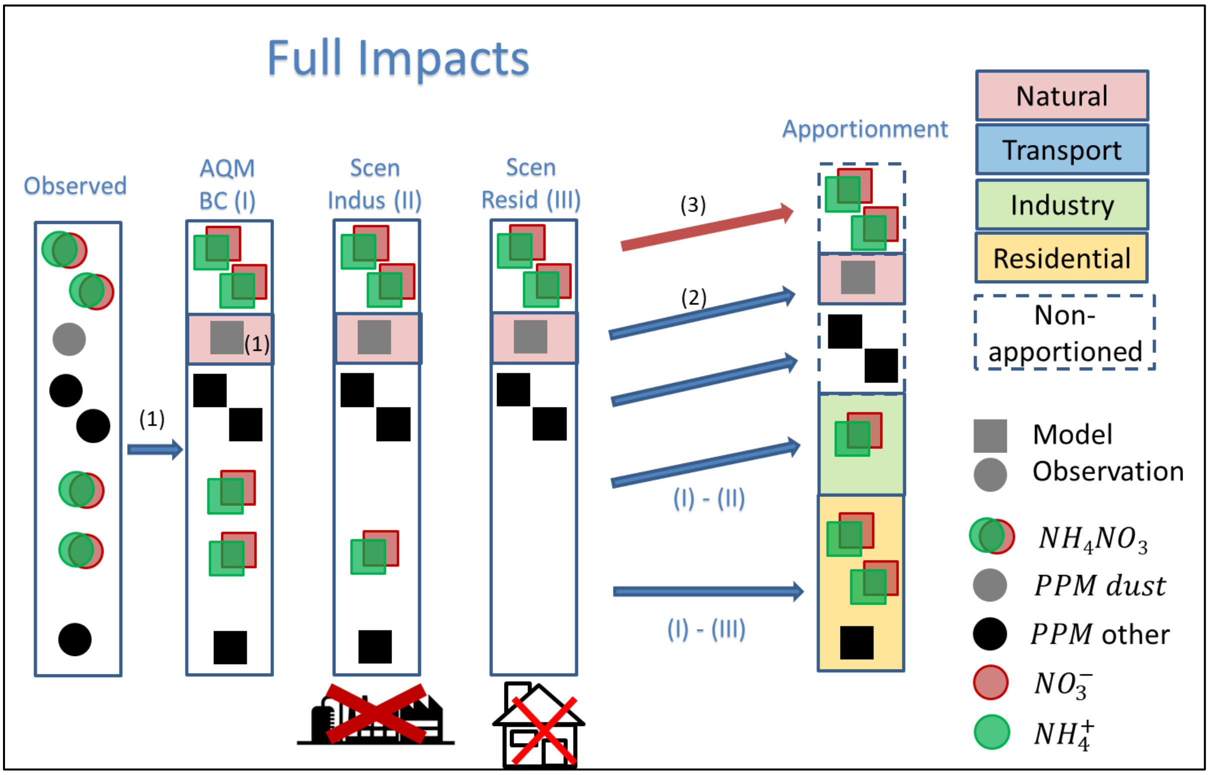

Figure 15: Process to obtain full impacts, starting from the measurements (left column) towards the final sectoral apportionment (right column). Sources that can be apportioned in terms of their sectoral origins are shaded with colours indicated in the top-right legend. Full impacts are model-based (arrow 1 towards columns (I-III) with squares)) and are obtained as the difference between a model base case (column (I) and a scenario (columns (II) or (III)) in which the source is removed. Full impacts do not apportion the species that are emitted outside the modelling domain (red arrows 3) 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 ((1) and arrow 2).

5.3.2 Source allocation

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

- Potential impacts are model-based (square symbols, columns (I-III)).
- 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 potential impact approach (pink shading - point 1 in Figure).
- The local industry and residential potential impacts are obtained as the differences between the modelled base case (column ((I)) and scenarios in which the local industry (column ((II)) and local residential (column ((III)) sectors are reduced. Given the 50% emission reduction applied, consistency and additivity are fulfilled and the potential impacts are obtained by multiplying these differences by a factor of 2. Background species are not apportioned (red arrows).

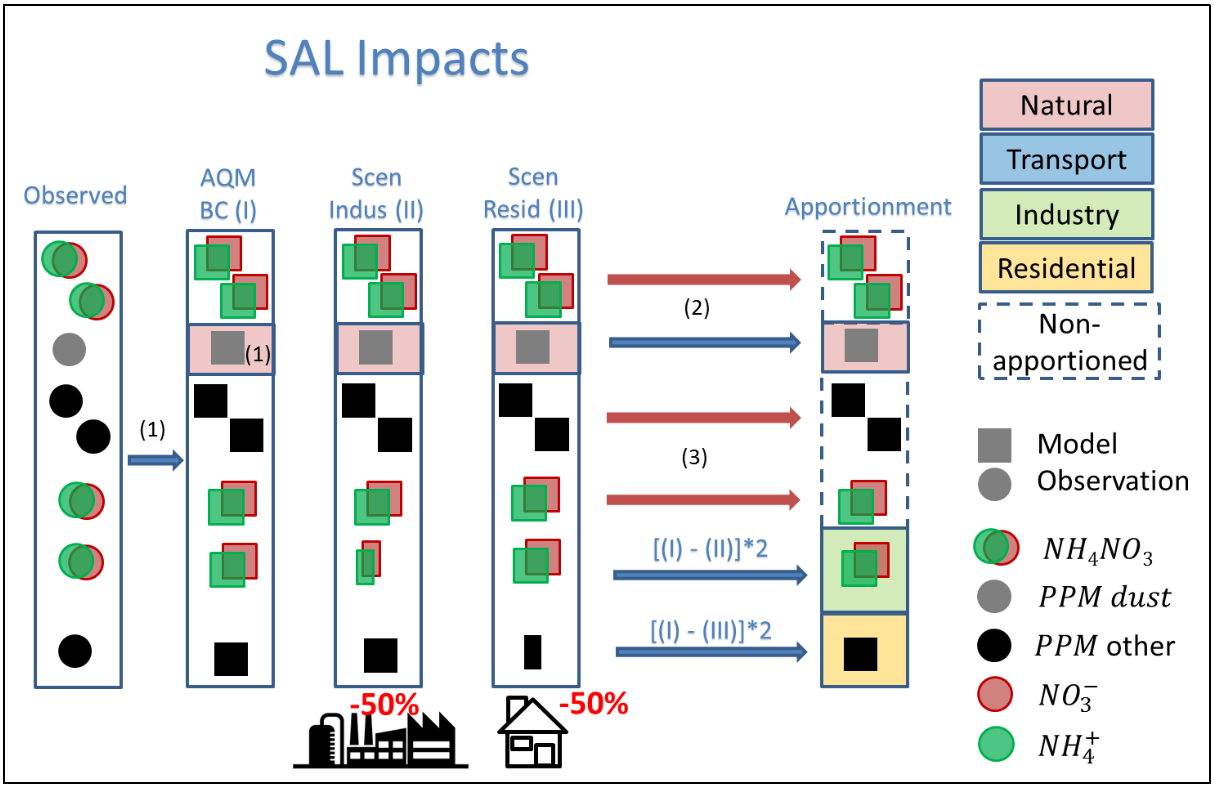

Figure 16: Process to obtain source allocation potential impacts, starting from the measurements (left column) towards the final sectoral apportionment (right column). Sources that can be apportioned in terms of their sectoral origins are shaded with colours indicated in the top-right legend. Source allocation potential impacts are model-based (arrow 1 towards columns (I-III) with squares) and are obtained as the scaled difference between a model base case (column ((I)) and scenarios (columns ((II, III)) in which the sources are reduced (here by 50%). Source allocation potential impacts do not apportion the species that are emitted outside the modelling domain (red arrows 3) 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 ((1) and arrow 2).

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 measurements.
- The background increment is equal to the measured background (column 2 from Figure 17).
- The local increment (3rd column) is obtained by subtracting the background increment from the observed concentration (1st column).
- From a sectoral perspective, the incremental approach does not deliver any information (right column), with the exception of dust (see first point above).

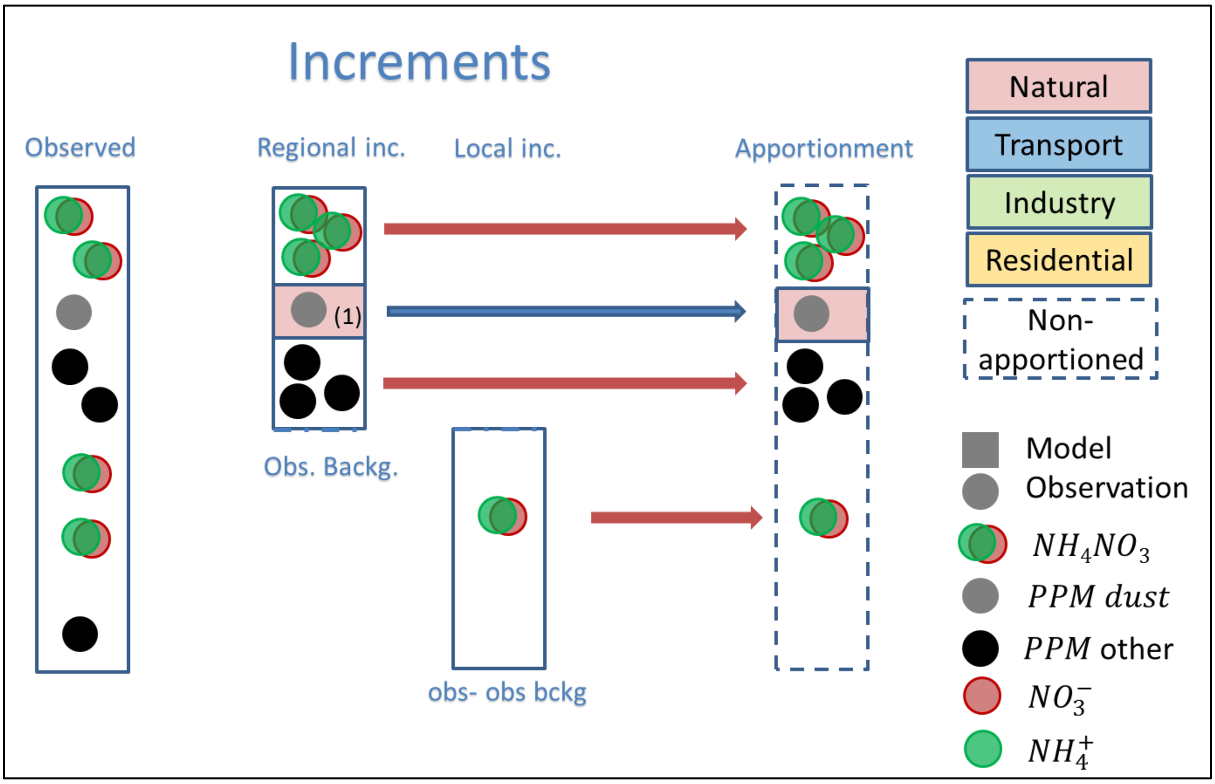

Figure 17: Process to obtain increments, starting from the measurements (left column) towards the final sectoral apportionment (right column). Sources that can be apportioned in terms of their sectoral origins are shaded with colours indicated in the top-right legend. Increments are measurement-based (circles). The background increment is equal to the measured background concentration (2nd column) while the local increment (3rd column) is obtained from the difference between the observations (1st column) and the background increment (2nd column). 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 (1).

5.5 Comparative overview

Sectoral source apportionment components as calculated from the different SA approaches applied to our example are summarised in Table 1 and Figure 18. We focus on the residential sector, although similar conclusions apply to the industrial sector.

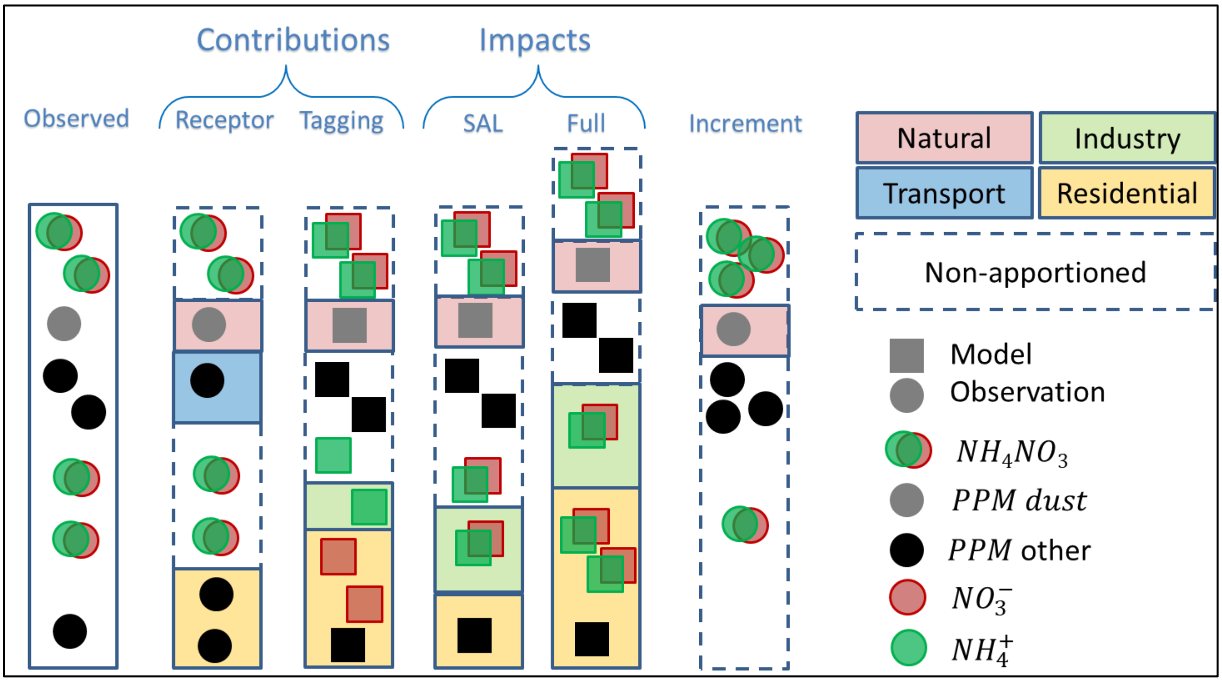

Figure 18: Graphical overview of the sectoral source apportionment components obtained with different approaches (SAL = source allocation). The results and differences shown here apply to the specific example described in Figure 12.

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.

	Residential component	**Industrial component**
Receptor contribution	● ●	None
Tagged contribution	■ 🟥 🟥	🟩
Full Impact	■ 🟩🟥 🟩🟥	🟩🟥
Source Alloc.	■	🟩🟥
Increment		

The comparative overview highlights the following differences:

- <u>Model vs. measurements</u>: in terms of sectoral apportionment, only the receptor model approach is measurement-based (circle symbols) while other methods are model-based. This implies that some information used by the former are inherently more correct and robust (e.g. bulk mass and chemical species concentration) than model-based approaches, where all information are derived from simulations. Conversely, source attribution with RMs mostly rely on statistical methods, while model-based approaches replicate physical and chemical processes underlying pollution formation, thus allowing a more transparent and robust analysis of the processes that link sources to atmospheric pollutant concentration. 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 sectoral apportionment, it is important to note that model-based approaches 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 corresponds to 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[1] (no red/green symbol); tagging only considers direct effects (only red symbols) while potential 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, potential impacts depend on the intensity of the emission reduction, as illustrated by the difference between full (5th columns, Figure 18) and source allocation (4th column).
- For a compound that is unique in terms of origin and that behaves linearly (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 sectoral apportionment.

In terms of model properties:

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 this prevents their use in supporting the design of air quality plans to manage non-linear species.

As full impacts arise from emission reductions, 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. Full impacts are indeed not additive, when non-linear species are involved.

Similarly, to the full impacts, source allocation impacts are dynamic and unambiguous by construction. Because they are calculated from a moderate enough range of emission reductions, source allocation potential 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 consistency and additivity are preserved.

Increments do not apply to sectoral apportionment.

These properties are summarised in Table 2.

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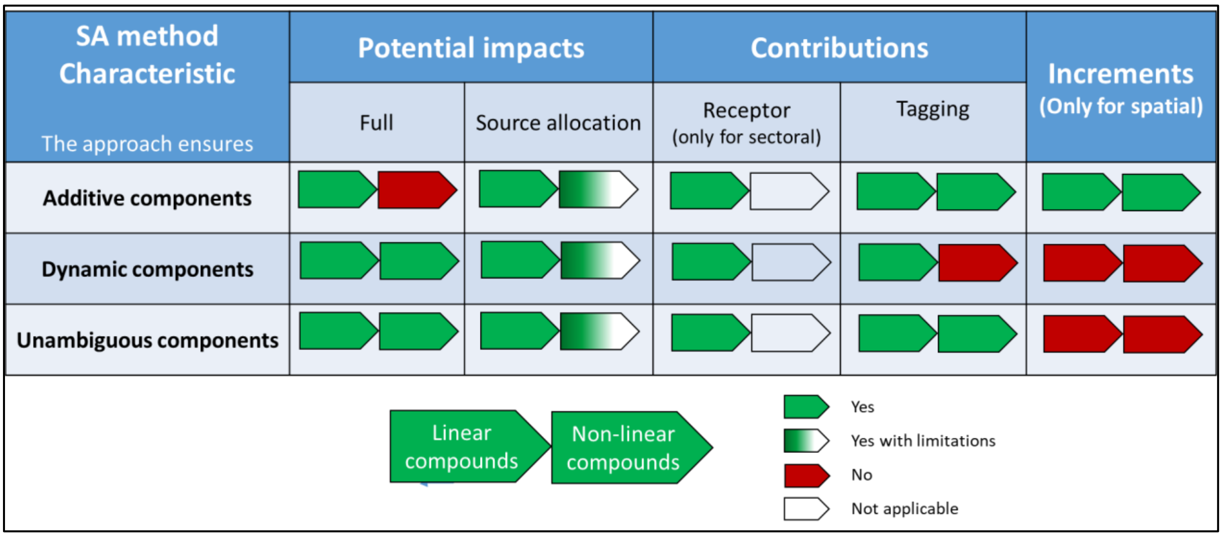
[1] For some non-linear species, specific markers can be used to identify a source

Table 2: Summary of properties attached to SA approaches when applied to linear and non-linear compounds. Note that receptor contributions only apply to sectoral apportionments whereas increments only apply to spatial apportionments. The gradient shading for the source allocation stress the fact that the approach only manages the non-linear compounds that are involved in linear processes (this occurs when applied for a limited range of emission reductions) and is not applicable beyond.



SA method Characteristic	Potential impacts		Contributions		Increments (Only for spatial)
The approach ensures	Full	Source allocation	Receptor (only for sectoral)	Tagging	
Additive components	🟩 🟥	🟩 ◧	🟩 ⬜	🟩 🟩	🟩 🟩
Dynamic components	🟩 🟩	🟩 ◧	🟩 ⬜	🟩 🟥	🟥 🟥
Unambiguous components	🟩 🟩	🟩 ◧	🟩 ⬜	🟩 🟩	🟥 🟥

Linear compounds | Non-linear compounds

🟩 Yes
◧ Yes with limitations
🟥 No
⬜ Not applicable

6 Spatial apportionment

We apply a similar reasoning for a spatial apportionment and use the same 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. Note that the final overview results shown below highlight differences that would remain valid for cases that are more complex (e.g. apportion different emitting regions).

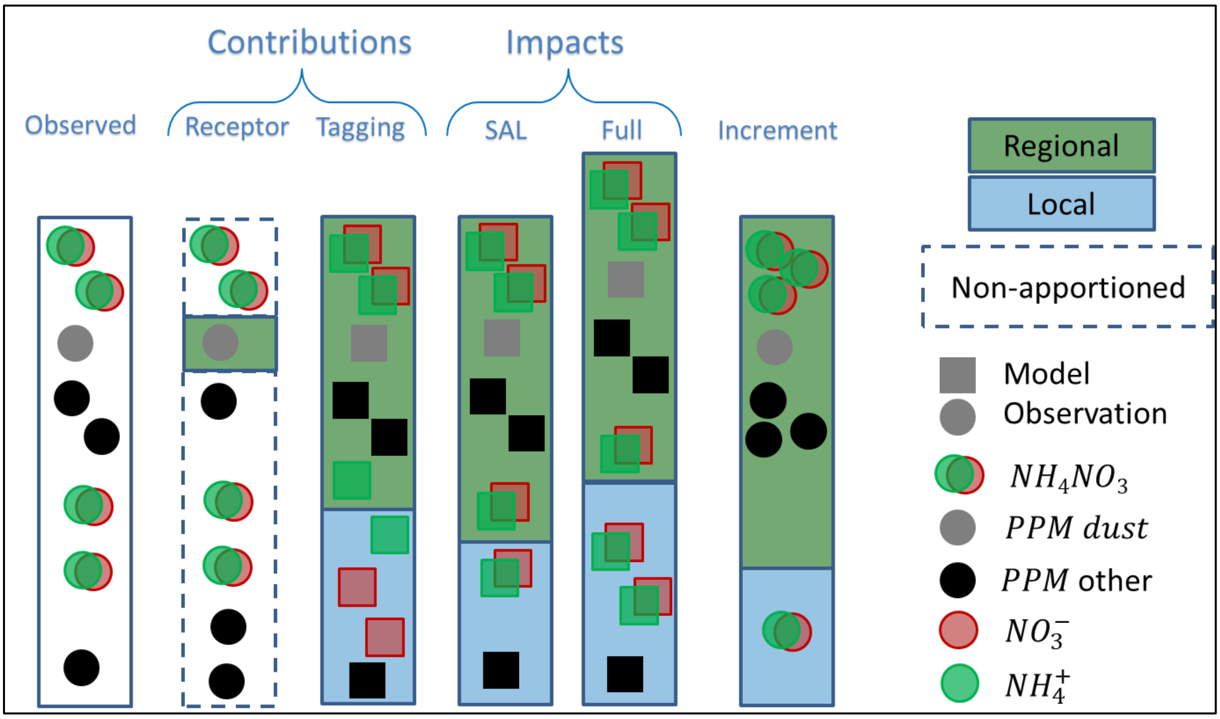
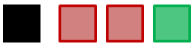


Figure 19: Graphical overview of the spatial source apportionment components obtained by different approaches (SAL = source allocation).

- By construction, receptor models (if used alone) are not able to perform a spatial apportionment and cannot therefore distinguish local from background source 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 sectoral fractions cannot be quantified.
- The local and background potential impacts are obtained by reducing the local emissions and the background, respectively and calculating the resulting difference on concentration. As shown by the full option (5th column, Figure 19), one of the main issues is the lack of additivity. Indeed the sum of the local and background potential impacts does not equal the baseline concentration. Additivity is only fulfilled for limited emission reductions as for source allocation (50% reduction in our case – 4th 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 3.

Table 3: Overview of the source apportionment components obtained by different approaches for spatial apportionment (based on illustrative example). See explanations for symbols in the text.

	Local component	**Background component**
Receptor contribution		
Tagged contribution		
Full Impact		
Source Alloc.		
Increment		

For *tagging contributions* and both types of impacts (*source allocation* and *full*), the properties attached to spatial SA are similar to those attached to sectoral apportionment. *Receptor contributions* are not applicable to spatial apportionment if used alone. Conversely, *increments* only apply to the spatial apportionment. They are additive by construction, however, the incremental components are unambiguously associated with the sources, only when two specific assumptions are fulfilled (see Section 4.2 and Annex). 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 (Table 2).

All source apportionment methodologies presented above are based on measurement and/or 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 potential 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 improve the accuracy and robustness of air quality modelling systems. With these two purposes in mind, this guide provides advice on how to use SA techniques in the overall context of air quality management. The use of SA to support 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 (Implementing Provisions on Reporting - 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.

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.

This implies that *for non-linear species*, only approaches that deliver *potential 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 proportional to the number of sectors/regions to consider), source allocation is the recommended approach.

For linear species, both the approaches that deliver *potential 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 recognised limitation for air quality planning applications.

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

As the pollutant formation processes become more non-linear, effective policies may not necessarily be those tackling the most dominant emission source but those tackling a substance that may be more scarce but has a critical role 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 simulations of various emission reduction scenarios will be able to support an effective policy strategy when non-linear processes are involved. Of course, even that approach has limitations due to 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 potential impacts-based approaches, not all sources behave non-linearly and particular conditions may apply to specific sources (mentioned in Table 4).

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

Source	**AAQD reference**	**Recommendation**
Transboundary pollution	Art. 1.20	As country emissions involve both linear and non-linear species, the potential impact-based approach is recommended.
Natural sources	Art. 20	A distinction must be made between linear (e.g. dust) and non-linear (e.g. NOx) species. For non-linear, only potential impacts are suited while for linear, measurements, contributions or potential 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 potential impacts. Given the uncertainty on the source strength and the well recognisable fingerprint, receptor modelling is however recommended.

With respect to the e-reporting of source apportionment (data flow I – see summary Table 5), potential impacts-based approaches are recommended given the non-linear nature of most sectors/sources, with the exception of the natural sources mentioned above. Indeed, most sectors (transport, industry etc.) emit significant amounts of compounds (e.g. NO_x, SO_2, NH_3 …) involved in non-linear chemical reactions. Used alone, receptor modelling approaches are not suited given their limitation to compounds behaving linearly as well as their inability to distinguish sources spatially (see Section 5.1 for details).

The incremental approach (used alone) is not suited given its inability to provide sectoral information.

As explained earlier, potential impacts-based methods, source allocation in this context, only provide information up to a given threshold for which consistency 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 an interest to use tagging contributions to complement potential impacts to provide more exhaustive information. This point is discussed in the "open issues" section. The table below summarises 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 (IPR), here focused on particulate matter.

Table 5: Recommendations regarding the use of SA approaches to produce e-reporting (Green=recommended; Red=not recommended; Orange=only as complementary information).

PM		Receptor contributions	Tagging/labelling Contributions	Source allocation	Increments
Background	Transboundary	To be complemented by impacts for the non-linear PM fraction	As complement of potential impacts?	🟩	🟥
	Country				
	Natural	🟩	🟥		
Urban	Traffic	To be complemented by impacts for the non-linear PM fraction	As complement of potential impacts?	🟩	
	Industry				
	Agriculture				
	Residential				
	Shipping				
	Off-road				
Local	Traffic				
	Industry				
	Agriculture				
	Residential				
	Shipping				
	Off-road				

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, potential 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

We begin with the example of a comparison between receptor and tagging/labelling contributions (a similar reasoning applies to the comparison between receptor contributions and potential impacts). From the illustrative example (Figure 18), we know that the main points that differentiate the results obtained with the two methods are the following (*potential issues - PI*):

PI1. Measurement vs. model: While receptor contributions are based on measurements, tagged contributions are model based (● vs. ■ in the illustrative example). This potential issue, as explained below, in specific conditions can be an opportunity to validate/verify emission inventory data or to identify sources not included in the model (e.g. ● from transport in the illustrative example).

PI2. 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, Figure 13 and Figure 14).

PI3. Non-linear fraction: Receptor models (RMs) do not allow performing 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, tagging algorithms can tag both primary and secondary compounds to a corresponding emission source (nothing vs. 🟥🟥 in the illustrative example).

Advice for the comparison: To overcome these potential issues it is necessary to ensure the following to retrieve valuable information from the inter-comparison:

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

The main benefit of the comparison is the comparison of model-based results with measurements (i.e. *model validation*). Although limited to linear compounds, this comparison can constitute a very good tool 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 types of compounds are correct.

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

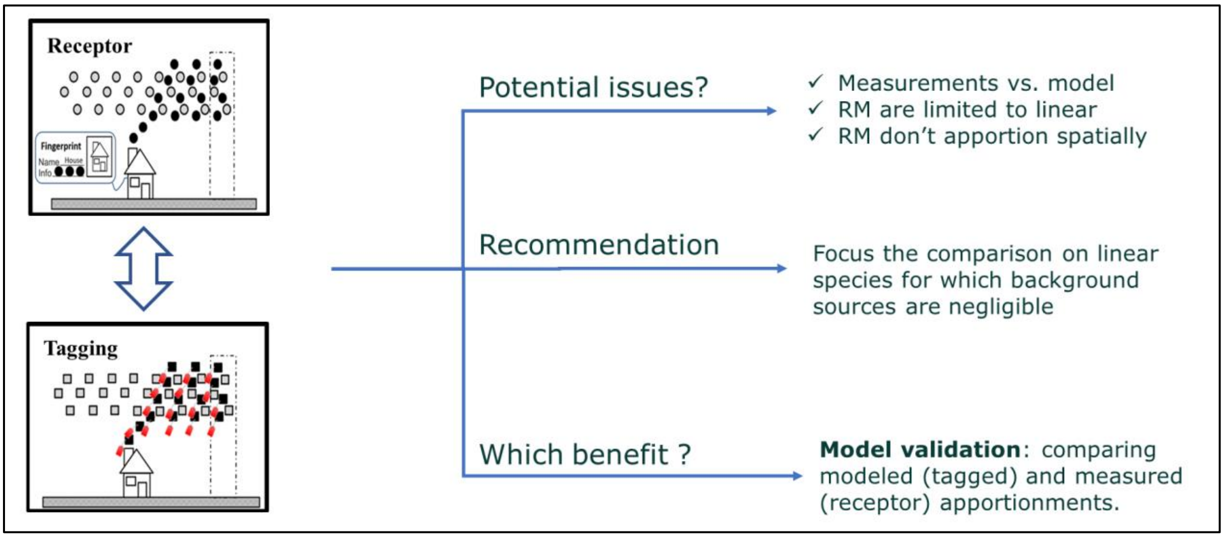


Figure 20: Synthetic overview of the potential issues (top right) met when receptor contributions (top illustration) are compared to tagging/labelling contributions (bottom illustration). The recommendation and potential benefit from the comparison are shown at the mid-right and bottom-right, respectively.

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 potential impacts is not recommended because of the following issue.

PI4. Increments suffer from a possible ambiguity, whose importance is impossible to assess (see the two incremental assumptions described in Annex A).

8.3 Potential impacts vs. tagging/labelling contributions

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

PI5. Direct/indirect effects: The main point that differentiates tagging/labelling contributions and potential 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. $NO_x \rightarrow NO_3^-$ and $NH_3 \rightarrow NH_4^+$) while potential impacts account for indirect effects (e.g. $NO_x \rightarrow NH_4^+$ or $NH_3 \rightarrow NO_3^-$).

PI6. Single vs. multiple apportionments: Each potential 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 <u>benefit of the comparison</u> is to improve model robustness.

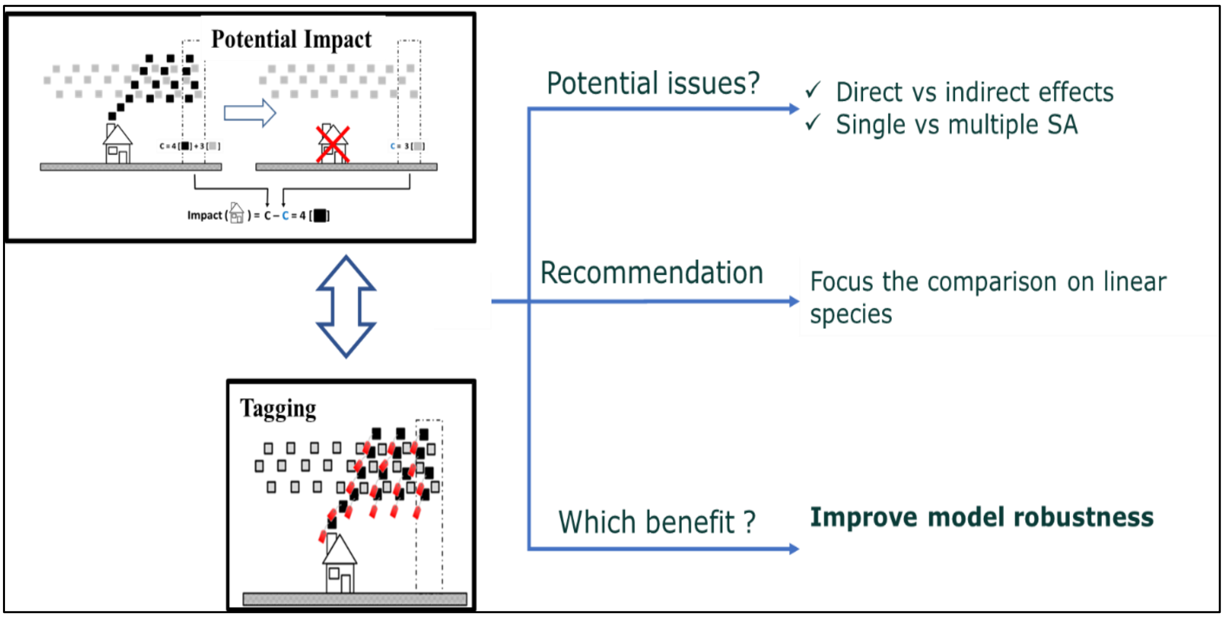


Figure 21: Synthetic overview of the potential issues (top right) met when potential impacts (top illustration) are compared to tagging/labelling contributions (bottom illustration). The recommendation and potential benefit from the comparison are shown at the mid-right and bottom-right, respectively.

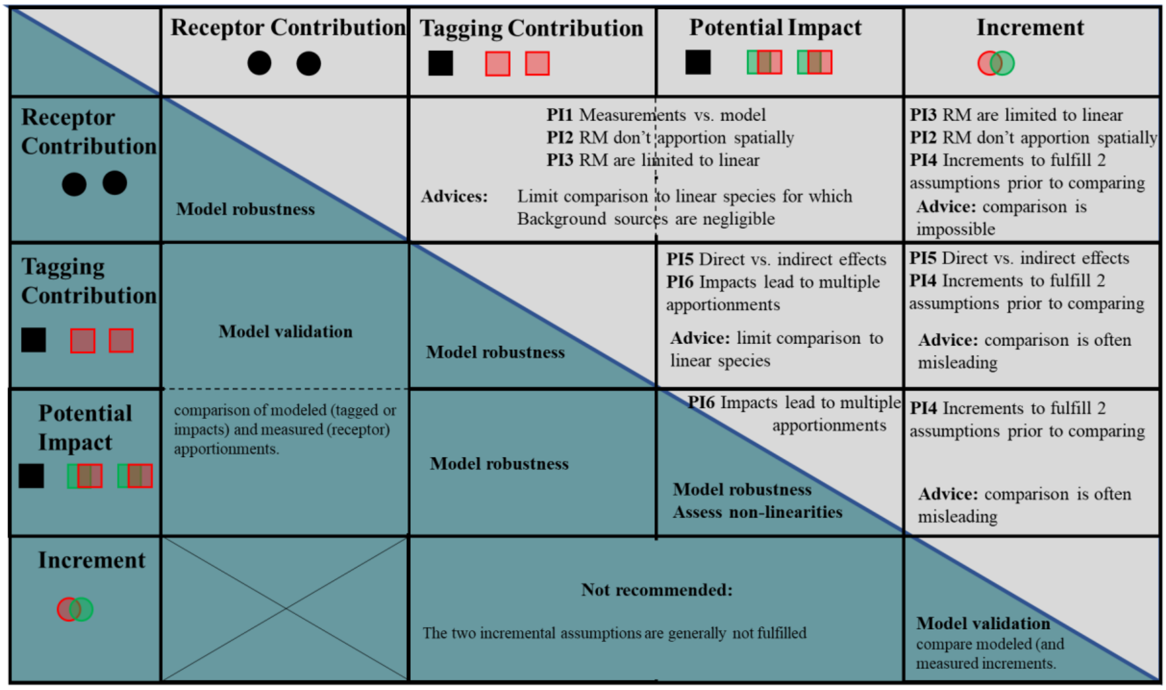
8.4 "Intra"-comparison of approaches

In general, there is no restriction for comparing methods of a similar type (e.g. tagging vs. tagging, increment vs. increment…). 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
- Comparison of potential impacts obtained with different strengths 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 summarised in Figure 22.



	Receptor Contribution ● ●	**Tagging Contribution** ■ 🟥 🟥	**Potential Impact** ■ 🟩🟥 🟩🟥	**Increment** 🔴🟢
Receptor Contribution ● ●	**Model robustness**	**PI1** Measurements vs. model **PI2** RM don't apportion spatially **PI3** RM are limited to linear **Advices:** Limit comparison to linear species for which Background sources are negligible		**PI3** RM are limited to linear **PI2** RM don't apportion spatially **PI4** Increments to fulfill 2 assumptions prior to comparing **Advice:** comparison is impossible
Tagging Contribution ■ 🟥 🟥	**Model validation**	**Model robustness**	**PI5** Direct vs. indirect effects **PI6** Impacts lead to multiple apportionments **Advice:** limit comparison to linear species	**PI5** Direct vs. indirect effects **PI4** Increments to fulfill 2 assumptions prior to comparing **Advice:** comparison is often misleading
Potential Impact ■ 🟩🟥 🟩🟥	comparison of modeled (tagged or impacts) and measured (receptor) apportionments.	**Model robustness**	**PI6** Impacts lead to multiple apportionments **Model robustness** **Assess non-linearities**	**PI4** Increments to fulfill 2 assumptions prior to comparing **Advice:** comparison is often misleading
Increment 🔴🟢	☒	**Not recommended:** The two incremental assumptions are generally not fulfilled		**Model validation** compare modeled (and measured increments.

Figure 22: The potential issues (and advice to address them) are detailed at each row/column intersection while the column/row intersection inform on recommendations for 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

9 Extension to other pollutants

The guide currently focuses on particulate matter (PM) because most of the methods are usually applied for this pollutant. Can we extend it to other species like NO_2 and O_3?

Questions:

- *Can we and do we have experience in using receptor models for* NO_2 *and/or* O_3*?*
- *Are there examples of using incremental methods for* NO_2 *and/or* O_3*?*
- *How can we translate our current recommendations on PM for* O_3 *or* NO_2 *for tagging methods?*
- *What are the consistency/additivity limits for source allocation for NO2 and/or O3?*
- *Can we extend the e-reporting table to O3 and or NO2?*

10 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. non-reactive 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 ammonium nitrate).

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?*

11 Use of RM to improve model based approaches: the case of organic aerosol (OA)

OA simulation remains a challenge for most AQM due to the uncertainties related to emission inventories (quantification of semi-volatile and intermediate volatility primary organic compounds), formation mechanisms (quantification of volatile precursors and formation yields), and atmospheric ageing. Still it accounts for a significant fraction of particulate matter, both in urban and rural environments, and air quality plans need to include it.

OA comprises hundreds of different chemical species with both primary and secondary origins. OA sources include industry, transport, residential, and biogenic emissions. During the last decade, SA based on high time resolution observations (RM) offered the opportunity to apportion organic aerosol mass to primary sources (including transport, wood burning, coal combustion) and secondary components, distinguished by their oxidation degree or based on the correlation with molecular markers of different sources (Canonaco et al. 2013; Srivastava et al., 2019). This approach supported the improvement of model performance in term of total OA mass and OA components (Jiang et al., 2019)

The main assumption of RM techniques is that each single aerosol source is characterised by a constant chemical profiles, or fingerprints, during the investigated time frame. This constraint does not strictly apply to most organic aerosol components. So a novel RM algorithm using a rolling window ME-2 approach was developed and implemented, to the capture seasonal variations of OA factors (Daellenbach et al., 2016).

Questions:

- Can RM results be used to improve OA description in SAL for air quality plans?

12 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. 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?

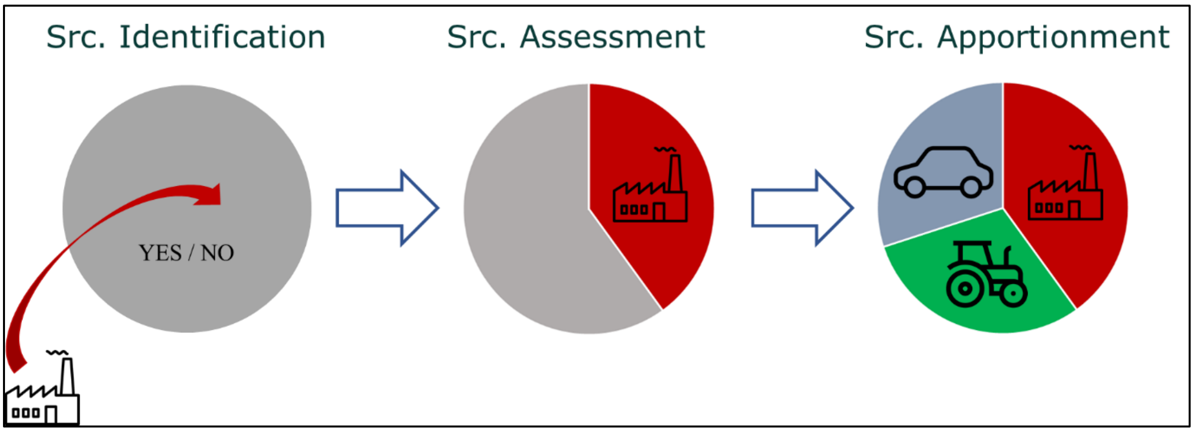


Figure 23: Schematic differences between source identification, assessment and apportionment

As an example, receptor models are somehow capable of "apportioning" certain types of non-linear secondary compounds (e.g. classes of organic aerosol - OA) indirectly via other related compounds (e.g. tracers) or via their properties (e.g. analysis of their oxidation degree or of correlation with molecular markers of different sources). This method allows one 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 sectoral 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 sectoral origins (e.g. wind-blown dust)? Can we detail for which sources this would be valid and how to proceed?*

13 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 potential 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 to emission reduction strengths up to 60% and provides for these levels a straightforward source apportionment that is dynamic, additive and consistent (see Section 4.5). This is however not the case beyond that emission reduction level.

For assessing direct policy measures (i.e. measures that involve reducing one sector at a time), full 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 potential impacts provide a top-down view on which actions (and associated reduction strength) will effectively reduce concentrations, tagging contributions provide a bottom-up view on the sectors to prioritise, when emission reductions go beyond a given threshold (either for direct or combined measures). It is interesting to note that while source allocation only targets industry in our example (because NH3 was the limiting compound), tagging contributions target all three sources as the chemical regime is not anymore NH3-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**.

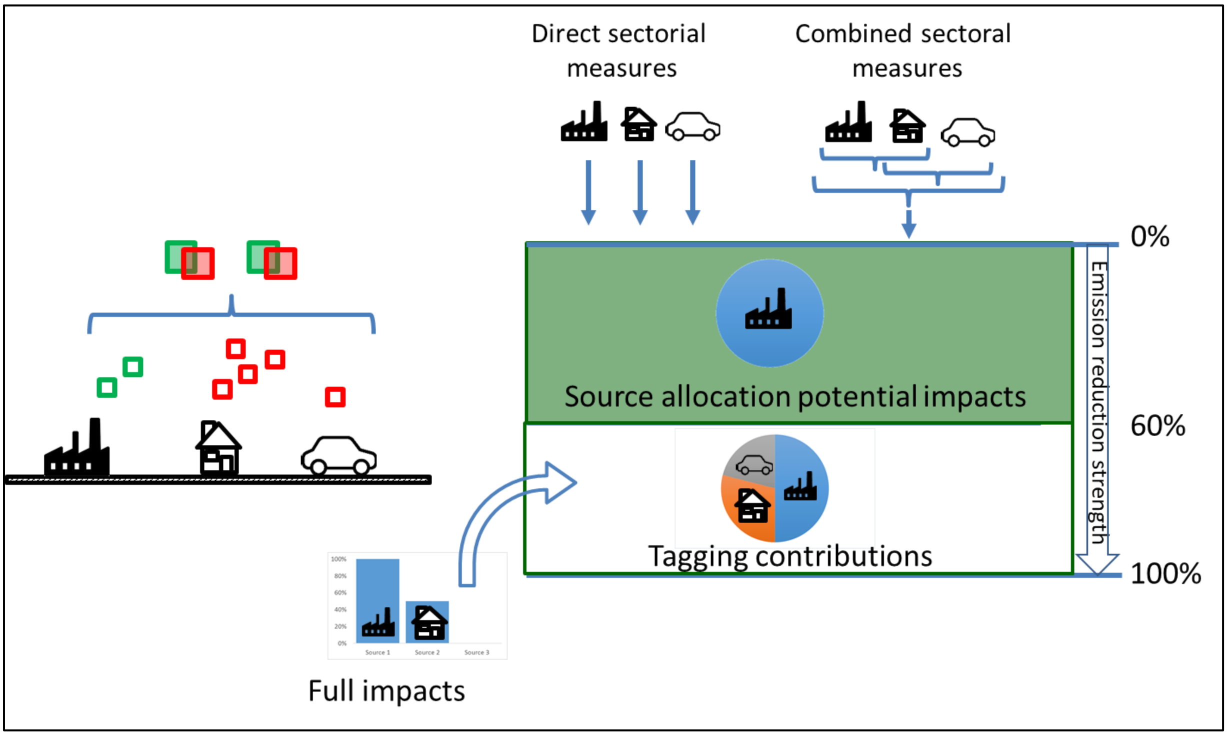


Figure 24: Three sources (left) emit either NH_3 (green) or NOx (red) compounds that combine on a 1:1 basis to form ammonium nitrate (combined solid green-red boxes). The right table differentiates direct (only one sector reduced at a time) from combined policy measures (more sectors are reduced contemporaneously). The

threshold of application of the source allocation is indicated by a horizontal line (here at 60%). The green shading indicates the zone where the applied method is consistent, additive and dynamic. For direct measures beyond the source allocation threshold, full impacts can be calculated but are not additive (sum of contributions > 100%).

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

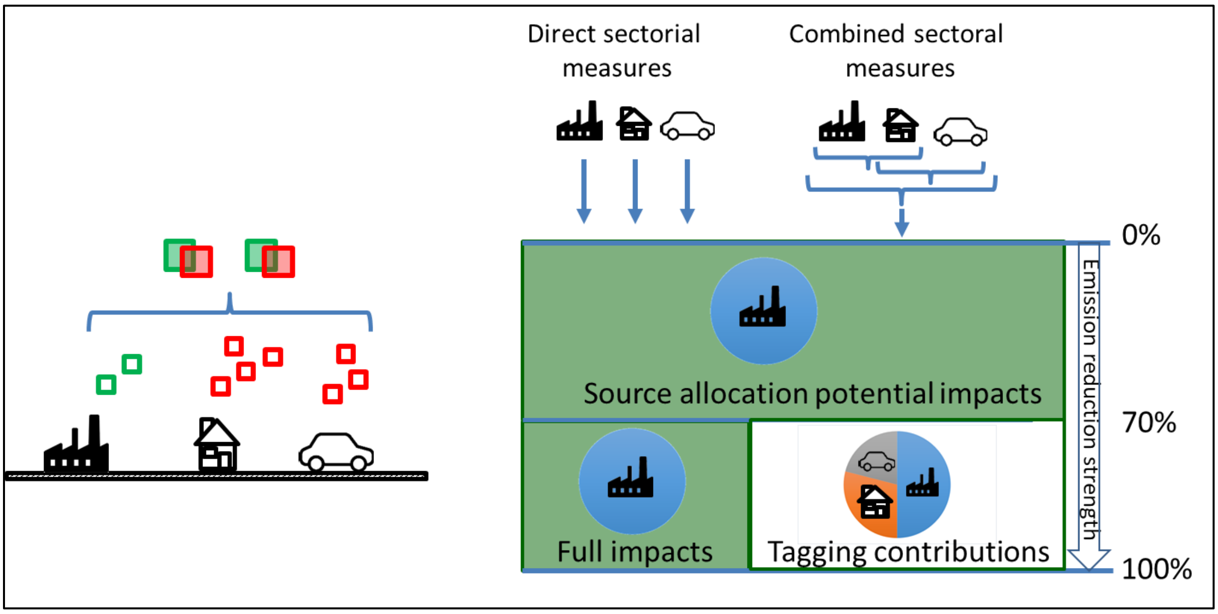


Figure 25: Three sources (left) emit either NH₃ (green) or NOx (red) compounds that combine on a 1:1 basis to form ammonium nitrate (combined solid green-red boxes). The right table differentiates direct (only one sector reduced at a time) from combined policy measures (more sectors are reduced contemporaneously). 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 consistent, additive and dynamic. For direct measures beyond the source allocation threshold, full impacts can be calculated and is additive (sum of contributions = 100%).

Question:

Potential impacts are dynamic 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 contributions as complementary information to the potential impacts to support planning? Potential 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)?

14 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?*

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Annexes

ANNEX 1. 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 ("u" superscript), the urban impact "I" is defined as:

$$I_{city}^{u} = C^{u} - B_{city}^{u} \qquad \text{(A1)}$$

where C^u is the concentration level reached at location "u" when both in-city and extra-city emissions are active and B_{city}^{u} is the background concentration level reached at the same location when city emissions are set to zero. This is represented in Figure 26.

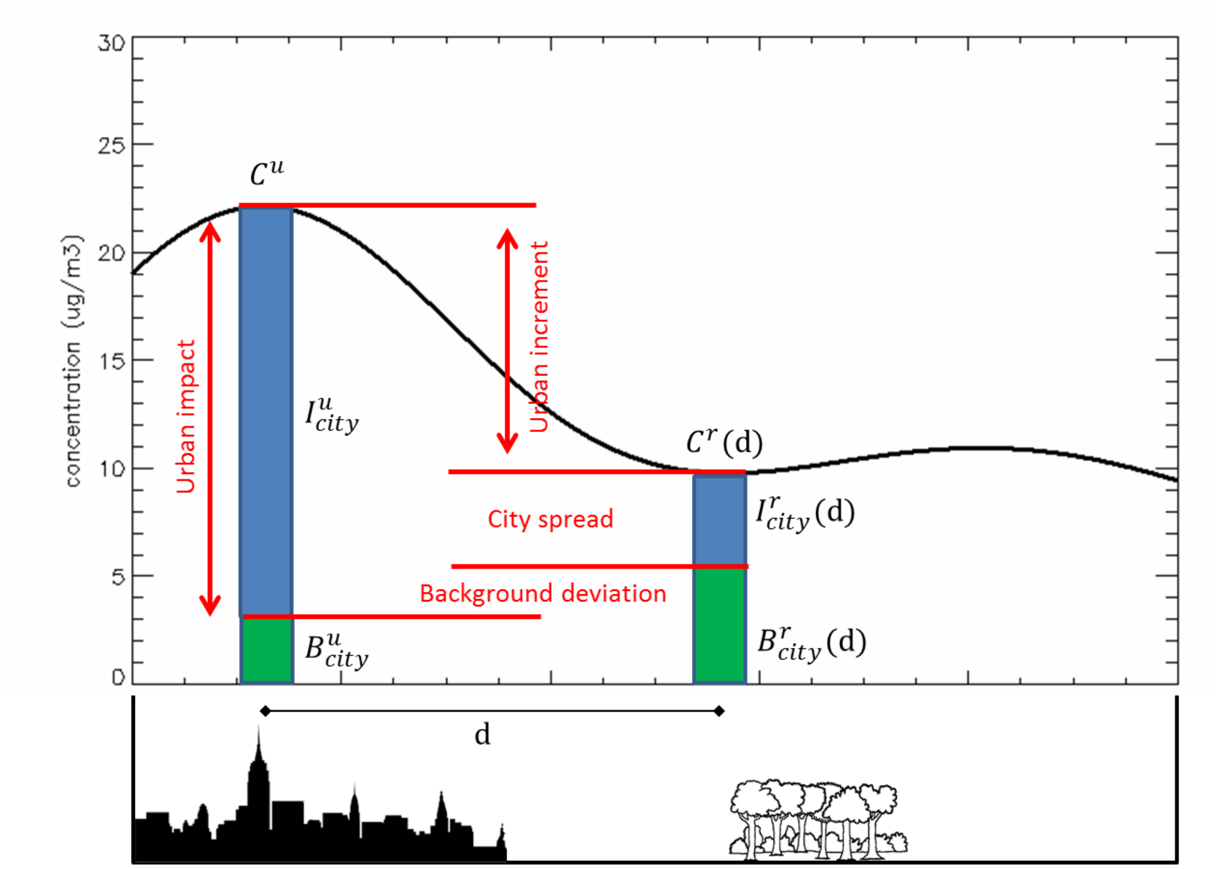

Figure 26: Schematic comparison of the urban impact with the urban increment. See text for details of the various components.

The urban impact (I_{city}^{u}) and the background (B_{city}^{u}) 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 the city emissions on the rural background location:

$$I^{r}_{city}(d) = C^{r}(d) - B^{r}_{city}(d) \qquad \text{(A2)}$$

The concentration at any location ($C^{r}(d)$) is then the sum of two components: an urban impact ($I^{r}_{city}(d)$) that represents the concentration due to the in-city emissions and a background level ($B^{r}_{city}(d)$) that represents the remaining concentration when in-city emissions are set to zero. At the city centre, the distance "d" equals 0 and Equation (A2) turns to Equation (A1)

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

$$I^{u}_{city} = \underbrace{(C^{u} - C^{r}(d))}_{Lenschow\ increment} + \underbrace{I^{r}_{city}(d)}_{City\ spread} + \underbrace{(B^{r}_{city}(d) - B^{u}_{city})}_{Background\ deviation} \quad \text{(A3)}$$

According to relation (A3), 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. We assume here isotropy but in practice, the urban increment also depends on the geographic orientation of the background station with respect to the city. Urban increments calculated at the same distance but with two different background stations are likely to differ, depending on factors like wind regimes, orography, land-use, etc.

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.

The incremental approach implicitly assumes that the city spread and background deviation terms can both be neglected at the distance (d) where the background measurement station is located, so that the impact of the city equals the Lenschow increment. Because these two assumptions are most often never met, the incremental approach is ambiguous. In other words, the urban and rural measurements are not perfectly representing the city and the rural background, respectively.

List of abbreviations and definitions

AQQD	Ambient Air Quality Directives
AQM	Air Quality Model
AQP	Air Quality Plan
EEA	European Environment Agency
EU	European Union
IPR	Implementing Provision Rules
ME-2	Multi-linear Engine 2
NH_3	Ammoniac
NH_{4+}	Ammonium
NH4NO3	Ammonium Nitrate
NO_3-	Nitrate
NO_x	Oxygen dioxide
O_3	Ozone
PI	Potential Impact
PPM	Primary Particulate Matter
PM	Particulate Matter
$PM_{2.5}$	Particulate matter with diameter less than 2.5 microns
RM	Receptor Models
SA	Source Apportionment
SAL	Source Allocation
SIA	Secondary Inorganic Aerosols
SOA	Secondary Organic Aerosols

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