Quantification of non-linearities as a function of time averaging in regional air quality modeling applications

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Abstract

Air quality models which are nowadays used for a wide range of scopes (i.e. assessment, forecast, planning) see their intrinsic complexity progressively increasing as better knowledge of the atmospheric chemistry processes is gained. As a result of this increased complexity potential non-linearities are implicitly and/or explicitly incorporated in the system. These non-linearities represent a key and challenging aspect of air quality modeling, especially to assess the robustness of the model responses. In this work the importance of non-linear effects in air quality modeling is quantified, especially as a function of time averaging. A methodology is proposed to decompose the concentration change resulting from an emission reduction over a given domain into its linear and non-linear contributions for each precursor as well as in the contribution resulting from the interactions among precursors. Simulations with the LOTOS-EUROS model have been performed by TNO over three regional geographical areas in Europe for this analysis. In all three regions the non-linear effects for PM10 and PM2.5 are shown to be relatively minor for yearly and monthly averages whereas they become significant for daily average values. For Ozone non-linearities become important already for monthly averages in some regions. An approach which explicitly deals with monthly variations seems therefore more appropriate for O3. In general non-linearities are more important at locations where concentrations are the lowest, i.e. at urban locations for O3 and at rural locations for PM10 and PM2.5. Finally the impact of spatial resolution (tested by comparing coarse and fine resolution simulations) on the degree of non-linearity has been shown to be minor as well. The conclusions developed here are model dependent and runs should be repeated with the particular model of interest but the proposed methodology allows with a limited number of runs to identify where efforts should be focused in order to include the relevant terms into a simplified surrogate model for integrated assessment purposes.

Keywords: Non-linearity, Air quality modeling, Photo-chemistry, Time averaging, Integrated assessment modeling, Surrogate models

1. Introduction

Air quality models are nowadays used for a wide range of scopes (i.e. assessment, forecast, planning) and see their intrinsic complexity progressively increasing as better knowledge of the atmospheric chemistry processes is gained and introduced in the model. While some decades ago models were mostly used to reproduce the ozone daily (or episodic) behavior their capabilities have now been extended and the same models are currently able to simulate the behavior of both gas- and particulate-phase compounds over extended time periods covering one or more years...
Thanks to these improvements, models now describe and simulate a more extended range of atmospheric processes. One of the side effects of this extended model capabilities is the implicit and explicit incorporation of non-linearities in the system. Indeed non-linear processes are included explicitly (e.g. chemistry) but interactions with pre-existing processes (e.g. feedback mechanism between meteorology and chemistry in on-line coupled models) can potentially occur as well and lead to implicit non-linearities. These non-linearities represent a key and challenging aspect of air quality modeling, especially when these models are used as a basis to provide information to policy makers.

One of the most useful applications of air quality models is indeed their possible use to assess the impact of emission abatement measures on air quality. This can be performed in a “scenario mode”, where the model is used to simulate both a base-case and an emission reduction scenario. Differences between the two can then be interpreted to deliver insight on the potential of selected emission reduction measures on air quality. This methodology can be used in practice to assess the impact of emission abatement measures at different temporal scales, ranging from few days to extended period of time covering one or more years (Cuvelier et al., 2007; Derwent et al., 2014). In all cases, it is crucial to know if the most efficient options (or emission reductions) resulting from the application of the air quality model are robust or not, i.e. if the model responses remain stable across the possible levels of emission reductions, and especially for levels of emission reductions which have not been explicitly tested, or for levels lying in between two tested levels of reduction. This is where non-linearities can potentially enter into play. Hence, information on the validity range of a linear assumption is important to save calculation effort when an optimal abatement scenario has to be determined.

The main objective of this work is to quantify the degree of non-linearity when air quality models are applied to assess the impact of emission reductions. This is the case for reductions of a single precursor (e.g. quantification of the degree of non-linearity of a NOx reduction on O3 by reducing NOx emissions by two levels, e.g. 20 and 50%) but also for potential non-linear interactions among precursor (i.e. the difference between reducing independently and together a set of emission precursors). To achieve this, we first introduce the methodology used to decompose the impact of emission reduction scenarios on concentration levels in terms of linear and non-linear components. Indicators are then constructed to quantify these various contributions.

In the second part of this section a description of the air quality modeling set-up is provided. It includes a brief description of the model and of the geographical application areas, focusing on the description of the emission scenario simulations performed in this study.

2. Methodology

The main objective of this work is to quantify the degree of non-linearity when air quality models are applied to assess the impact of emission reductions. This methodology is then applied on practical cases based on a set of model simulations performed in different locations within Europe. The objective of this work is to use the above mentioned methodology to:

- Quantify the degree of non-linearity as a function of the time averaging process and answer the question: Are longer time-averages reducing sufficiently the degree of non-linearity to allow a linear assumption when constructing the source-receptor model?
- Assess the dependencies of non-linearities in terms of geographical location (three different areas in Europe are tested).
- Assess the dependencies of non-linearities in terms of pollutant (O3 and PM) and precursors (NOx, VOC, NH3, PPM and SO2)
- Assess the impact of spatial resolution on the degree of non-linearity (two resolutions (7 and 28 km) are tested)

These topics are addressed in Section 3. Conclusions are proposed in Section 4.

2.1. Factor separation methodology and selected indicators

The methodology used in this work builds on the factor separation methodology proposed by Stein and Alpert (1993), extended to the specific case of emission reduction impacts by Thunis and Clappier (2014). In order to assess the importance of non-linear effects in air quality model applications, an indicator based on concentration delta (i.e. the difference between a base case and an emission reduction scenario) is defined. The concentration delta resulting from a simulation in which all precursors (denoted by the letter k) are reduced simultaneously by a percentage $\alpha$ (referred to as $\Delta C^{(k)}_\alpha$) can be decomposed in the following way:

$$\Delta C^{(k)}_\alpha = \sum_k \Delta C^k_\alpha + \Delta C^{int}_\alpha$$

where $\Delta C^k_\alpha$ are the concentration delta resulting from a reduction of one precursor at a time and $\Delta C^{int}_\alpha$ is the remaining contribution arising from the non-linear interactions among all precursors. The individual concentration deltas can further be decomposed into their linear and non-linear components as follows:
\[
\Delta C^{(k)}_a = \sum_k \Delta C^{k,\text{lin}}_a + \sum_k \Delta C^{k,\text{nlin}}_a + \Delta C^{\text{int}}_a
\]

\[
= \sum_k \frac{\alpha}{\beta} \Delta C^k_a + \sum_k \Delta C^{k,\text{nlin}}_a + \Delta C^{\text{int}}_a
\]

(2)

where \( \beta \) is an intermediate level of emission reduction between the base case and the \( a \) emission reduction scenario (i.e. \( 0 \leq \beta \leq a \)).

One of the main drawbacks of this formulation is that delta 

concentrations are unbounded and are not easily comparable among species. We therefore transform equation (2) in terms of relative change of concentration at one species. The single precursor linear and non-linear potencies, \( p_{k,\text{lin}}^a \) and \( p_{k,\text{nlin}}^a \), can be calculated similarly so that equation (2) now reads:

\[
p^{(k)}_a = \frac{\Delta C^{(k)}_a / C}{\Delta E^{(k)}_a / E} = \frac{\Delta C^{(k)}_a}{\alpha C}
\]

with the last equality resulting from the fact that the level of emission reduction for each precursor is comparable (\( \Delta E^{(k)}_a = \alpha E \)).

The single precursor linear and non-linear potencies, \( p_{k,\text{lin}}^a \) and \( p_{k,\text{nlin}}^a \), can be transformed similarly so that equation (2) now reads:

\[
p^{(k)}_a = \sum_k p_{k,\text{lin}}^a + \sum_k p_{k,\text{nlin}}^a - \sum_k p_{k,\text{int}}^a = \sum_k p_{k}^b + \sum_k p_{k,\text{nlin}}^a + p_{\text{int}}^a
\]

(3)

Relative potencies are usually bounded between \( -1 \) and \( 1 \), with a positive value indicating a concentration increase resulting from an emission reduction. As an example a relative potency of \(-0.5\) means that reducing emissions by 100% in the selected area will lead to a reduction of the concentrations of 50% at the selected location. When model responses are linear the single precursor potencies are constant (\( p_{k,\text{lin}}^a = p_{k,\text{lin}}^b \)) and the interaction potency becomes zero (\( p_{\text{int}}^a = 0 \)). More details on the construction and properties of the relative potencies can be found in Thunis and Clappier (2014).

The relative potencies of interest in this work are the non-linear potencies \( p_{k,\text{nlin}}^a \) for each precursor as well as the interaction potency \( p_{\text{int}}^a \) which can be calculated from known quantities as:

\[
p_{k,\text{nlin}}^a = p_{k}^b - p_{k}^a
\]

\[
p_{\text{int}} = p_{(k)}^a - \sum_k p_{k}^a
\]

In the following section a quantification of these terms is provided for different air quality model applications. For convenience we will refer to “precursor full potency”, “single precursor non-linear potency” and “interaction potency” for \( p_{k,\text{lin}}^a \), \( p_{k,\text{nlin}}^a \) and \( p_{\text{int}}^a \), respectively in the remaining of this work. All potencies can also be calculated for different time-averaged concentrations (i.e. day, month or year) but no additional symbol will be used. For convenience, we will rather refer to these different time-averaging’s explicitly during the discussion of the results. Before doing so, we first describe briefly the air quality model used in this work as well as the set of model simulations used in each of the selected geographical areas.

2.2. Model set-up and emission scenarios

In the current work, simulations performed by TNO with the air quality chemistry transport model LOTOS-EUROS (Manders et al., 2009) have been used over different geographical areas in Europe: Lombardy (Italy), 24,000 km² for 10 million inhabitants; Flanders (Belgium), 13,500 km² for 6 million inhabitants and Silesia (Poland), 12,300 km² for 5 million inhabitants (see Fig. 1). All simulations cover the entire meteorological year 2009. Since PM10
and PM2.5 depend on 5 different precursors (NOx, VOC, PPM, NH3 and SO2) the computation of the relative potency indicators described in the previous section requires a series of 12 independent simulations over each of the three geographical areas, i.e.:

- A reference simulation which will provide base case concentrations.
- One simulation with a reference level (a) emission reduction. This run is repeated for the 5 precursors. In the current work a has been set to 50%. This level represents a compromise between (1) a large enough reduction to capture the main aspects of the model responses to significant changes in the input data and (2) a level of reduction which remains realistically achievable in terms of human activity constraints and prevents the appearance of strange model behaviors (e.g. responses of models used in extreme parameter ranges).
- One simulation with an intermediate emission reduction level (β). This run is repeated for the 5 precursors. In the current work β has been set to 20%.
- One simulation in which all 5 precursor emissions are reduced contemporarily by the percentage a.

These twelve simulations are performed in three different geographical areas mentioned above to test potentially different chemical regimes. In all three cases, the emissions outside the domain are kept at base-case level. In addition these simulations are performed with both 7 and 28 km spatial resolutions to test the impact of spatial resolution on the degree of non-linearity.

3. Results and discussion

3.1. Full potencies as a function of time-averaging

Since a time behavior analysis for the potencies cannot be performed in practice for every grid points, three locations have been selected within each of the three simulation domains (Silesia, Flanders and Lombardy). In particular, to reflect the potential differences in terms of chemical regimes, an urban, suburban and rural station types have been selected in each domain: Antwerp

Fig. 2. Full PM10 potencies: For each location (X axis) the yearly (red asterisks), monthly (orange) and daily (black) full potency range are shown. The green shaded area indicates the ± 5% threshold, arbitrarily chosen to indicate negligible values. The cities considered are Antwerpen (An), Mol (Mo), Zandhoven (Za), Milan (Mi), Gallarate (Ga), Ispra (Is), Katowice (Ka), Myszkow (My) and Siewierz (Si). See text for more details. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)
as not-significant in terms of concentration levels, the potency is considered of 100%) within the considered domain leads to less than a 5% levels. In other words when switching off the emissions (reduction arbitrarily chosen to demark important from negligible potency indicative of potencies lower than 5%, a threshold which has been values and facilitate the visualization. The green shaded area is over 98 percent of the available potency values to avoid extreme and 3). For daily values, the maximum and minimum are calculated minimum values are kept (end points of the vertical bars in Figs. 2 calculated each month and day, respectively and the maximum and respectively. For monthly and daily averages, potencies have been calculated for all precursors reduces contemporarily are calculated on a daily, monthly and yearly basis. Figs. 2 and 3 show how these potencies vary in terms of precursors, geographical area, type of locations at these sites are used in the analysis, monitoring sites are selected because they are locations of special interest. At all 9 locations the potencies for each individual precursor as well as the potency for all precursors reduced contemporarily are calculated on a daily, monthly and yearly basis. Figs. 2 and 3 show how these potencies vary in terms of precursors, geographical area, type of locations and time averaging period for PM10, PM2.5 and O3 respectively. For monthly and daily averages, potencies have been calculated each month and day, respectively and the maximum and minimum values are kept (end points of the vertical bars in Figs. 2 and 3). For daily values, the maximum and minimum are calculated over 98 percent of the available potency values to avoid extreme values and facilitate the visualization. The green shaded area is indicative of potencies lower than 5%, a threshold which has been arbitrarily chosen to demark important from negligible potency levels. In other words when switching off the emissions (reduction of 100%) within the considered domain leads to less than a 5% change in terms of concentration levels, the potency is considered as not-significant.

For PM10, the following points can be highlighted:

- The span between the maximum and minimum daily potencies (length of the line) is considerable for all precursor potencies pointing out to the high variability in terms of model responses across days within the year. This variability is obviously reduced for monthly values although it remains significant. This also implies that model responses obtained on few day episodes cannot easily be generalized over a longer time-frame and vice-versa.
- Although some daily potencies are slightly positive, most of them are negative. Yearly and monthly averaged potencies provide negative values and, consequently, can be considered as adequate average representations of the range of daily values.
- Slight positive potencies are observed in some geographical areas for some precursors (i.e. an emission reduction of one precursor leads to an increase of concentration). This is especially visible for NOx in Lombardy and SO2 in the Silesia region.
- The potency resulting from the simultaneous reduction of all precursors \( p_{\text{all}} \) is generally dominating the single precursor potencies \( p_{\text{precursor}} \).
- Among the single precursor potencies, PPM and NH3 are the dominating precursors whereas the NOx impact is only significant in the Lombardy area.
- Although the model responses are relatively similar for Flanders and Silesia, significant variability is seen in terms of geographical areas. In terms of yearly averaged potency, the maximum potential for local action is seen in Lombardy, followed by Silesia and Flanders. Note that the characteristics in terms of area and population among the three regions could explain some of these differences.
- While the full potencies are generally higher at urban locations for O3, the reverse is true for their non-linear components which tend to be larger at rural stations. For PM10 and PM2.5 no clear trend can be identified.

For PM2.5 (not shown) the same conclusions can be made although potencies are generally slightly more important. This could be due to the higher fraction of biogenic (and/or dust, seasalt…) included in the primary coarse particulate fraction, leading to lower changes on PM10 concentration levels.

For O3 and in particular the daily 8 h maximum O3 average (Fig. 3), the NOx precursor full potencies are positive and important in all three regions whereas VOC potencies are weaker and negative. As expected no potency arising from SO2 and NH3 precursor reduction are seen (not shown). Similarly to PM10 and PM2.5 the span between the maximum and minimum daily potencies is considerable for all precursors, implying that model responses obtained on few day episodes cannot easily be generalized over a longer time-frame. While the monthly averaged potencies capture both the negative and positive range of values and therefore...
provide an adequate representation of the possible chemical regimes, this is not the case for the yearly average which only captures a slightly positive response. This might indicate a need to consider at least seasonally or monthly averages for O₃ to better capture the range of possible responses.

As a conclusion it is interesting to note the high variability of the potencies among the days of the year. In some regions like Lombardy, days with almost zero potency follow days with almost full potencies (not shown). The monthly and daily averaged potencies obviously capture only a small fraction of this variability.

### 3.2. Non-linear potencies as a function of time-averaging

We focus in this section on the non-linear components of the potencies for each pollutant (PM₁₀, PM₂.₅, and O₃) as a function of time-averaging. Regarding PM₁₀ (Fig. 4) the dominating non-linear component is the interaction contribution, although the Lombardy area also exhibits significant non-linearities for NOₓ and NH₃. While the range of daily non-linearities sometimes overshots the 5% threshold limit, this never happens to the monthly and yearly averages, with the exception of Lombardy where the monthly non-linear interaction term remains significant. As discussed in the following section, these important interaction non-linearities occur mostly during the summer time period.

As a conclusion a linear approach based on yearly averaged PM₁₀ values would be valid for the three geographical areas covered in this study. It is interesting to note that while the full potencies (section 3.1) are mostly negative (i.e an emission reduction leads to a concentration decrease), the non-linearities are quite balanced and exhibit a quasi-similar distribution between negative and positive values. Note also that non-linearities tend to be more important at rural and suburban locations, where the PM concentration levels are generally the lowest.

Non-linearities are slightly more important for PM₂.₅, most probably resulting from the dominating non-linear reaction mechanism responsible for the secondary formation of the fine particulate fraction. Here again the only locations where the monthly non-linearities exceed 5% belong to Lombardy where values exceed 20% during the summer time. In general a linear simplification approach for yearly averaged PM₂.₅ concentrations

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**Fig. 4.** Non-linear PM₁₀ potencies for the NOₓ (top left), SOₓ (top middle), VOC (top right), PPM (bottom left) and NH₃ (bottom middle) precursors and for the interaction term (bottom right): For each location (X axis) the yearly (red asterisks), monthly (orange) and daily (black) full potency range are shown. The green shaded area indicates the ± 5% threshold, arbitrarily chosen to indicate negligible values. The cities considered are Antwerpen (An), Mol (Mo), Zandhoven (Za), Milan (Mi), Gallarate (Ga), Ispra (Is), Katowice (Ka), Myszkow (My) and Siewierz (Si). See text for more details. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)
would also be valid although in Lombardy the yearly average non-linearity is slightly above the 5% threshold (around 7%).

For O₃ (Fig. 5) most of the non-linearities are seen for the NOₓ precursor in all three regions. In contrast with the other regions Lombardy also exhibit important non-linearities for the interaction term. While the VOC and interaction non-linearities are balanced between negative and positive values, the NOₓ non-linearities are predominantly positive.

Yearly averages exhibit very low level of non-linearities but they tend to capture very little of the daily variability. Monthly values exhibit quite important non-linearity levels, especially in the Lombardy region. As detailed in the following section, these daily and monthly NOₓ non-linearities are mostly positive during winter time and negative during summer time. It is also interesting to note that non-linearities tend to be more important at urban location, where the VOC limited regime is more pronounced, and O₃ concentrations generally lower.

In conclusions yearly averaged non-linearities are generally not important for all pollutants in the three regions tested in this work. But they become important for monthly and daily averaged concentrations and cannot anymore be neglected.

In Fig. 6 the full and non-linear potencies are shown for all three pollutants as a function of time. To facilitate the visualization, only the monthly averaged potencies for the maximum impact precursor are shown (i.e. the case in which all precursors are reduced contemporarily for PM and the case in which NOₓ only is reduced for O₃). As can be seen from Fig. 5 only one out of the three regions (Lombardy) exhibits a strong seasonality for PM₁₀ both in terms of the full potency and of its non-linear component. If we look at how this seasonality is distributed across the different precursors in Lombardy (not shown) the NOₓ and NH₃ precursor potencies show maximum (i.e. high negative) values in the late summer whereas the PPM potency peaks (i.e. high negative) in the winter time. Almost no seasonal variation is seen for SO₂. In the two other regional areas, Silesia and Flanders, no significant seasonal variations are seen. The same observations remain valid for PM₂.₅.

Regarding non-linearities, only one region (Lombardy) shows strong seasonal dependencies but these are only important for the interaction terms, i.e. arising from the complex interactions among precursors. These non-linearities peak during the summer time where they reach 15%.

For O₃ the same conclusions can be drawn. Only one region, Lombardy, shows important seasonal variations in terms of non-linearities which arise mostly from the NOₓ precursor contributions. These non-linearities are positive during the winter time and negative during the summer time. Since the non-linear potencies follow a similar pattern to the full potencies a linear modeling approach would therefore tend to underestimate the impact of emission reductions (both in positive and negative terms) throughout the year.

As discussed in the previous section an approach based on yearly averages for the maximum 8 h daily O₃ might miss some of the possible model responses and in particular miss the negative regime. From the results discussed in this section an approach based on seasonal averages would already solve most of this drawback. It is also important to note the strong dependency on location, suggesting a specific approach for each particular case.

3.3. Full and non-linear potencies as a function of concentration levels

Similarly to the previous section, we limit our analysis to the monthly averaged potencies for visualization purposes. Fig. 7 (top panel) shows the full potencies at the 9 locations as a function of O₃ concentration level for the two precursors NOₓ, VOC and their interaction. As can be seen the NOₓ potency tends to dominate and is largely positive at most locations for the low and medium range of concentration levels. Only at the high end of the concentration range (occurring mostly at suburban and/or rural station) does the NOₓ potency become slightly negative, indicating a potential for a reduction of the O₃ concentration levels. As discussed in the previous section, these negative potencies occur mainly during the summer time. The VOC potencies are always negative but tend to show larger values at high concentration levels. The potential for reducing O₃ levels from either NOₓ or VOC precursors emission abatement seems to be limited to a few months only.

The non-linear components follow a similar trend but with much more negative values than for the full potencies. These values

![Fig. 5. Non-linear O₃ daily 8 h maximum potencies for the NOₓ (left) and VOC (middle) precursors and for the interaction term (right). For each location (X axis) the yearly (red asterisks), monthly (orange) and daily (black) full potency range are shown. The green shaded area indicates the ± 5% threshold, arbitrarily chosen to indicate negligible values. The cities considered are Antwerpen (An), Mol (Mo), Zandhoven (Za), Milan (Mi), Gallarate (Ga), Ispra (Is), Katowice (Ka), Myszkow (My) and Siewierz (Si). See text for more details. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)](image-url)
become important however only at the higher and lower range of concentrations. A linear approach would again tend to underestimate the increase of $O_3$ concentrations at low concentration levels and the decrease of $O_3$ concentrations at high levels. The nonlinearities related to the VOC precursor remain unimportant over the whole range of concentrations at all locations considered in this work. It is also interesting to note that for both the full and non-linear potencies the interaction term (right figure of Fig. 7) has a very similar pattern to the NOx potency one.

In contrast to $O_3$, the potencies (full and non-linear) only show limited trends with respect to the concentration level for PM10 (Fig. 8).

### 3.4. Precursor potencies and non-linearity's as a function of spatial resolution

All full and non-linear potencies discussed in the previous sections have been obtained on the basis of model simulations performed at 7 km resolution. In this section we test the impact of model spatial resolution by repeating all simulations with a 28 km spatial resolution. Regarding PM10 full potencies (Fig. 9) very little differences are seen between the two spatial resolutions. With the exception of the rural station in Silesia, the results obtained with 28 km resolution tend to underestimate slightly the 7 km potencies. This is especially clear for all Lombardy locations. The 28 km resolution would therefore lead to very similar full potency responses to the 7 km resolution simulations. The same conclusions hold for PM2.5. The non-linear potencies exhibit the same behavior and the differences in terms of non-linearity between the 7 and 28 km resolution potencies are not significant.

For the ozone 8 h daily maximum (Fig. 9), the impact of resolution on the full potencies is more visible, especially at urban locations where the decrease in resolution (28 km) tends to shorten the range of positive NOx potencies significantly, especially for daily averages. No differences are seen for yearly averaged potencies and only slight ones for monthly averaged values. Similar conclusions can be drawn for the non-linear component of the potencies. As a reminder these non-linearities become only important for monthly

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**Fig. 6.** Full (top) and non-linear (bottom) monthly average potencies for daily averaged PM10 (left), daily averaged PM2.5 (middle) and the 8 h daily maximum O3 (right) as a function of time of the year for all 9 locations. The two horizontal lines indicate the ±5% threshold, arbitrarily chosen to indicate negligible values. Lombardy cities are in green, Silesia in red and Flanders in blue. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)
and daily averaged concentrations and for monthly averages this is the case only in Lombardy. The move to a coarser resolution affects mostly the urban locations but the differences remain minor. More differences are seen in terms of daily values.

3.5. Precursor potencies and non-linearity's as a function of the emission reduction domain size

One of the possible drawbacks of the methodology used in this work is the possible dependency of the results on the choice of the domain over which emissions are reduced. Since the relative potency is directly proportional to the concentration delta, one would then guess that this delta could be proportional to the emission reduction domain. To test this hypothesis a series of independent simulations has been performed in which emissions have been reduced over the whole European territory. The potencies are calculated at the same nine locations with both 7 and 28 km spatial resolutions. The full potencies are obviously much larger (not shown) but the main question is to know whether their non-linear component will also be more important.

These non-linear potencies are shown in Figs. 10 and 11. For PM$_{10}$ slight differences can be seen for the single precursor potencies but these changes remain small and the yearly and monthly averaged non-linear potencies remain within the 5% significance threshold. This is not true for the interaction component which now becomes important and ranges between 5 and 15% for yearly averages and approaches 25% for monthly values. These interactions therefore become important for an emission reduction domain which size varies between a region (no impact in the cases tested above) and the whole European domain (extreme case tested here).

The same conclusions remain valid for PM$_{2.5}$ although the observed potencies are generally larger (in absolute terms).

For the O$_3$ 8 h daily maximum, the yearly averaged non-linear potencies remain below the 5% significance threshold for both precursors (NO$_x$ and VOC) as well as for the interaction terms. No real difference is seen resulting from the choice of the domain over which emission are reduced.
4. Conclusions

The main objective of this work was to quantify the importance of non-linear effects in air quality modeling, especially as a function of time averaging. While non-linear effects have been shown to be significant for hourly or daily averaged results, it was still uncertain how much of these non-linear effects would remain when results are averaged on a monthly or yearly basis. Since this time average process is commonly assumed in integrated assessment modeling to produce simplified source-receptor models, it is important to assess the remaining degree of non-linearity in order to guarantee that these source receptor surrogate models deliver robust solutions.

In order to achieve this objective, a methodology building on Stein and Alpert (1993) and Thunis and Clappier (2014) has been proposed to decompose the concentration change resulting from an emission reduction over a given domain into its different components. In particular the linear and non-linear contributions for each precursor are identified as well as the contribution resulting from the interactions among precursors.

A series of simulations was performed over three regional geographical areas in Europe and in each of these areas; three locations (urban, suburban and rural) have been chosen to assess the importance of non-linear effects as a function of time averaging.

For both PM$_{10}$ and PM$_{2.5}$ the full potencies are generally dominated by the interaction term and are mostly negative (i.e. a reduction of emission leads to a reduction of the concentration levels). For these two species the monthly and yearly average potency therefore summarize quite adequately the daily variabilities. In all three regions the non-linear effects are relatively minor for yearly and monthly averages although in one region (Lombardy) some non-linear effects were shown to become important for monthly averaged values. In the latter region the non-linear effects show a strong seasonality with negative potency values during the summer time, whereas in the other two regions no strong seasonal dependency was observed.

For Ozone (8 h averaged daily maximum value) the full potencies are dominated by the NO$_x$ precursor contribution which
shows large positive values during the winter time (characterized by relatively low concentrations) and slightly negative values during the summer time (characterized by relatively high concentrations). While the monthly averaged potencies capture these different regimes, it is not the case for the yearly average slightly positive potency which misses to capture the negative range of the potencies. Due to these possible differences in terms of chemical regime throughout the season it might be recommendable to use at least seasonal indicators for O₃ (in place of yearly averaged ones). All regions show a seasonal dependency regarding the full potency but only one, Lombardy, shows this dependency for the non-linear potencies.

It is interesting to note that the full and non-linear potencies have generally a similar shape and sign for PM₁₀ and PM₂.₅ indicating that a linear simplifying assumption would lead to underestimating the impact of any emission reduction measure. For ozone the same conclusions do not hold as the non-linear potencies are not following so straightforwardly the full potencies in terms of shape and sign.

Another point worth to mention is the tendency of having larger non-linearities at locations where concentrations are the lowest, i.e. at urban locations for O₃ and at rural locations for PM₁₀ and PM₂.₅. If the scope of modeling is the assessment of limit values exceedances, one might allow a larger error (by neglecting non-linear terms) at locations were concentrations are relatively smaller or far from the exceedance threshold.

Similar simulations have been performed with 7 and 28 km spatial resolution to check whether a coarser resolution would tend to increase/decrease the importance of the non-linear terms. In general very minor differences have been found either for the full and non-linear components of the potencies.

Finally the impact of the dimension of the geographical area over which the emissions are reduced has been tested by considering the extreme cases in which emissions would be reduced over the entire Europe. While for PM₁₀ and PM₂.₅ some significant differences are seen and might lead to the need to consider monthly average values in place of the yearly average, this is not the case for O₃ where the conclusions remain valid regardless of the dimension of the emission reduction domain.

Although air quality models generally show a robust behavior in terms of responses to emission changes (van Loon et al., 2007) differences do exist among models in terms of amplitude and level of non-linearities characterizing these responses (Pernigotti et al., 2013; Cuvelier et al., 2007). The conclusions developed here

Fig. 9. Comparison of daily, monthly and yearly potencies obtained with 7 km and 28 km (square symbols). Daily averaged PM₁₀ and PM₂.₅ potencies are on the left and middle, respectively while the 8 h daily maximum O₃ potencies are on the right. Full potencies are on top and non-linear potencies at bottom. Other figure details are similar to Fig. 2.
Fig. 10. Same as Fig. 4 for PM10 but full (top) and non-linear (bottom) potencies are obtained with emissions reduced over the entire Europe (not only on the regional domain, as in Fig. 2).

Fig. 11. Same as Fig. 5 for the O3 8 h daily maximum but full (top) and non-linear (bottom) potencies are obtained with emissions reduced over the entire Europe (not only on the regional domain, as in Fig. 2).
therefore remain model and region dependent. The proposed methodology allows however with a limited number of runs to inter-compare model responses among each other but also to identify where efforts should be focused in order to include the relevant terms into a simplified surrogate model for integrated assessment purposes.

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References


