

We thank the two referees for their constructive and pertinent comments.

We have now significantly extended the level of detail in the text, and have been more explicit on different points, like the requirements for an implementation in a new software and the tool's limitation to linear chemistry.

Below is a point by point answer to the specific comments. In blue are our comments, in black the original referee comments (first RC1, then RC2, and the revised manuscript with colors showing the modifications at the end).

Specific comments RC1

- First of all the authors claim that their method is inexpensive wrt. the computational costs. Considering Table 1 I agree that the costs are low for small window sizes (e.g. 11x11 or 21x21), but with larger sizes the costs get remarkably large. Of course a simulation with 161x161 would replace 161x161 perturbation simulations (if I understand the method correctly), but for source apportionment studies one would usually not be interested in the contribution from 161x161 gridboxes to each other. Instead, one would be more interested in contributions from different regions/and/or emission sectors, right? For linear species this could be achieved by “just” adding additional tracers for different regions/emission sectors which should end up in far less additional costs compared to the large overhead for a 161x161 window. To better judge the advantages of the method the authors should make clear how large the windows (x,y and z direction) for different purposes need to be. For the downscaling example given in Sect. 4.2 or the example in Sect. 4.3 small windows might be enough, but for the source apportionment I guess that very large windows are necessary. At least from the information of Figs 5,6,7 and 8 I have the feeling that 9 levels and a window of 80x80 are necessary (at minimum) to account for all sources (e.g. a completeness of ≈ 1). Further, pollutants with different lifetimes should need different window sizes (for a completeness of 1, see Fig.4). I think it is very important that the authors characterise the method in more detail and critically discuss the advantages and disadvantages of their method compared to other methods for different scientific questions.

Yes, the method may indeed be very expensive for some applications. We have tried to be more careful about our claims. If the main goal is to get country-to-country source receptor matrices, then it may not be the right tool. We thus have added in section 5 “If the goal is to provide source-receptor matrices for large regions (countries), then this method is probably not appropriate in its present form as the computational cost may be too high, and the level of detail provided is not needed. For such an application the method should be modified, so that the tracking is not done for individual grid cells, but for larger source areas or group of emission sources.”

- In the description of the method the authors explain how they deal with emissions, advection, diffusion, deposition and chemistry, but it remains unclear how to deal with convection. On the coarse grid applied in the example convection needs to be parametrized I guess (providing convective mass fluxes). Therefore, please describe how you deal with convection in the LF framework.

Yes, this should be commented. In section 2.3 we have now added

“For convection the same procedure can be used by replacing the diffusion operator in Eq. (11) by the convection operator. In the current EMEP MSC-W model version, the convective processes are not implemented in the Local Fraction calculations.”

And further in section 3:

“The standard settings of the model do not include convection over Europe.”

- The presentation of the examples is rather vague and many details are missing. Clearly, the main goal of the authors is the description of the method in general (e.g., the mathematical concept), which is fine. However, the results depends heavily on the implementation of the method in one specific model and the authors give no details about the implementation procedure in EMEP MSC-W. The authors claim that 'The updates of the Local Fractions can be added on top of an existing model in separate routines.' (p14|7ff) I am not familiar with the EMEP model, but when inspecting the provided code it seems to me that several modules (including the advection module) needed to be touched to implement the method. Therefore, I would strongly recommend that the authors add more details about the technical implementation in the model (maybe as Supplement). In this context I ask the authors to please provide examples of CTMs/CCMs where the flux at each grid box is an available quantity. To my opinion, the flux might not be an easily available quantity in many models (depending heavily on the applied advection scheme).

We have added section 4 with an explicit description of what is needed from an implementation point of view.

- The authors are not providing any details of the model set-up. Of course the manuscript is not intended as model evaluation etc., but some basic details of the set-up would help to understand the presented results and it would help other users of EMEP to reproduce the results.

We added "The parameter settings are essentially the same as what is used for the official EMEP-MSCW runs, using "TNO_MACC-III" emissions (2015 update of (Kuenen et al., 2014)). However to simplify the interpretation of the results, two important modifications have been introduced: a simplified advection scheme is used (see Sec. 3.2), and all emissions are released at the lowest level."

- The method assumes linearity for chemical processes, which is acknowledged by the authors. However, the authors state in the abstract that the method is valid for all primary pollutants, even though some of them have non-linear chemistry (e.g. NO_x, NO₃). This should be clarified in the abstract. Further, the authors discuss in Sect. 4.4 that they aim to include chemical processes. The consideration of the full chemistry, however, would lead to a highly more complex implementation (and more costly wrt to the computational resources). In this case 'traditional' tagging approaches (e.g. Li et al., 2012, Emmons et al., 2012, Kwok et al., 2015, Valverde et al., 2016, Grewe et al., 2017, Butler et al. 2018 (and references therein)) might be superior compared to the LF approach. Could the authors please comment on this?

As mentioned in the abstract "non-linear chemical processes are not accounted for and only primary pollutants can be addressed", and the details of what the term "not accounted for" means for primary pollutants which are non-linear, is defined in section 2.5: the method will give some results, but the stronger the non-linear effects are, the less accurate the results will be. Also, in the intro we mentioned that "It will thus complement existing methods, but not replace them"

Nevertheless, we have modified the last section: "In the future we plan to generalize the method to also include chemical processes in some simplified form. The ambition is to still provide information for a very large number of sources, but to describe chemical processes in an approximate way. Compared to existing tagging methods, it will trade accuracy for computational efficiency."

- On p14|11ff & p15|3 the authors mention the large memory footprint of the method. Information about the relative increase of the memory demand caused by the LFs would be interesting. How much more nodes are needed if the method is applied?

We have added an entry in Table 1, showing memory demand. (The number of nodes

depends on the amount of memory available on each of them. 64 GB per node or more is typically available on modern HPC systems, meaning that the demand is not unreasonably large for the window sizes considered here)

- The definition of 's' is unclear in the manuscript. On p317 's' is defined as the source term (i.e. emission sector). On p6125 's' is defined as pollutant. In all example there is no differentiation between emission sectors and I assume that the number of emission sectors is 1 in all examples? Information like this should be part of the model description Section.

Conceptually, as far as the Local Fraction is concerned, there are not much differences between sectors and pollutants. They are just different sources. In fact, our implementation does use the same index for describing both.

- Please reconsider the usage of the term 'contribution'. Especially in the introduction (p2111ff) 'contributions' and 'impacts' are mixed. With the 'direct' method only impacts can be calculated and only for linear species 'impact' equals 'contribution' (see e.g., Thunis et al., 2019).

Modified to "difference"

Technical corrections:

- The colour bars in Figure 4 are not very helpful. They are showing many orders of magnitude, which make it hard to judge the difference between direct and LF. Maybe additional difference plots could be provided additionally.

Yes, but this large range makes it also difficult to plot meaningful relative or absolute errors.

The point here is to show that the shape of the distribution looks the same, also for non-linear species. To quantify the limits would be interesting, but would require too many different test cases.

This must be done in an other study.

- Please check the references. For example Emmons et al., 2012 is listed in the bibliography but the reference seems to be missing in the manuscript.

The list of references has been checked and revised

- Further, I do not see how a reference to a manuscript in preparation is useful in any way (Denby et al.). This is recommended by the journal ("Works "submitted to", "in preparation", "in review", or only available as preprint should also be included in the reference list"). We hope to be able to add more details in the reference before publication. Even without more details, it should help future readers to search for it, once both papers are published, hopefully.

- p2120f: Why using present perfect here? I would say the tagging method is unpractical in these cases.

Modified to

"In cases where the number of different tagged sources is large this method becomes unpractical."

- Caption figure 2: Please clarify what total emissions averaged over on month are. Are you showing total emissions of the month or an monthly average flux of molecules?

This was not clear in the manuscript; wording now changed to "accumulated".

Specific comments RC2

Page 1, line 4: "distinguish a large" should probably be "distinguish the contribution of a large".

Yes, this has been changed.

Page 2, line 17: The references given here are not a good representation of studies which trace pollutant concentration back to emitted species by tagging (with or without consideration of nonlinearities). For PM, the study of Kranenburg et al. (2013) should be mentioned here. It is mentioned later in the manuscript, but should also be discussed here because it is a tagging method. For ozone, both Wang et al. (1998) and Wu et al. (2011) actually avoid the nonlinearities by tagging ozone based on its geographical region of formation, rather than the geographical region in which precursors are emitted. The reference to Grewe et al. (2013) is more appropriate here, since it is capable of attributing ozone to its emissions, but the authors could consider instead referencing the most up-to-date version of this method, as described by Grewe et al. (2017) and an actual application of this method for ozone attribution by Mertens et al. (2018). Alternative approaches also exist, which make different trade-offs. The approaches shared by Dunker et al. (2002) and Kwok et al. (2015), which tag ozone based on the chemical regime are both well-established and should be referenced in any discussion of modelled source attribution. Yet another approach was described recently by Butler et al. (2018) and has been applied by Lupascu et al. (2019). The authors are right that large number of tracers can rapidly make tagging computationally expensive, but they should also point out that techniques exist to keep these problems in check. For example Butler et al. (2018) Restrict the number of tracers to a carefully chosen set of representative source sectors; Grewe et al. (2017) make use of the concept of "chemical families" to keep the number of tagged species within reasonable limits; and Lupascu et al. (2019) restrict the length of their simulation period to focus on a pollution episode of interest. Each of these approaches brings different trade-offs, but in each case also significant advantages: the ability to perform source attribution for secondary pollutants; and the ability to perform source attribution for long-range transport. These trade-offs are especially interesting in the context of the present manuscript, since one of the major ways in which the computational complexity of the local fractions method is kept computationally simple is by restricting the size of the "local region" for which the source attribution is performed.

Thank you, we have included the recommended references.

However since our method only considers primary pollutants and exclude (at present) ozone, we prefer not to discuss in too much detail the additional challenges involved.

Yes, it is important to relate our method to other methods showing the trade offs involved and which type of applications where the Local Fraction method has its advantages (and the ones where it is not recommended). We have included a new paragraph in the Discussion section.

Page 2, line 33: "can be built ... relatively easily" is a vague statement. More detail is needed here.

We have added a new section about implementation with a more explicit description of what is needed and why we think it is "relatively easy to implement". In the introduction we have added "In Sect. 4 we will give an overview over what is required to implement the method in an existing CTM and discuss the performance in the EMEP MSC-W implementation."

Page 3, line 5: An important detail missing here is that origins of the pollutants being tracked must be restricted to emissions within a "local region". This should be made clear up front, rather than making the reader wait 2 more sub-sections to find this out. Yes, we have added a new paragraph in the intro (see below)

Page 3, line 8: "source regions" is very vague here. It would help the reader to know already at this stage that the present implementation considers each grid cell as a separate source region, but that in principle the method can be expanded to work with larger source regions. Yes, we have added a new paragraph in the intro (see below)

Page 4, lines 7-8: "reasonable" cost and "preset" numbers of grid cells are used very vaguely here. These terms are discussed later in the manuscript, but most readers would benefit from forward references to the relevant sections here.

Yes, we have added a new paragraph in the intro (see below)

Page 4, lines 12-13: "usually not necessary..." is very vague here. The authors show later that in fact extending vertical resolution to at least the height of the PBL is useful. The authors should also note that this also applies only to the pollutants they assess in their manuscript. Transport in the free troposphere is important for some pollutants such as ozone.

Yes, we have added a new paragraph in the intro (see below)

Applications that need an explicit description of transport to the free troposphere (and back) would require more vertical levels. Therefore we have clarified that the size of the local region must be adapted to the specific needs of the application.

Page 4, line 17: Keeping the size of the LF array down to a reasonable size appears to be the main trade-off associated with this method, and this should be acknowledged here.

We have also added an entry in Table 1 that show quantitatively what this means.

Page 4, line 18: The concept of the "local region" is first used here, and only implicitly defined by its context. It would help most readers tremendously if this concept could be introduced a lot earlier, with the explanation that setting the size of the local region represents the major trade-off with using this method.

(Thank you, such things are difficult to realize for authors, but important for new readers!)

We have added a new paragraph in the introduction:

“In principle the method allows the definition of any group of sources, but here we will show results only for the case where each defined source is defined within a single grid cell. One key limitation, which makes the method manageable, is that the tagged values are stored only up to a preset horizontal and vertical distance from their source. We will call the region within this distance the "local region". The size of this region must be set as a balance between computational cost and the accuracy requirement of the application.”

Page 5, line 20: This is a good point, and could perhaps be mentioned earlier in the manuscript where the authors make the claim that their method is easy to implement in different CTMs.

Page 6, lines 8-9: Generally throughout the manuscript it would also be nice to have some discussion of the limitations of the method.

Page 6, line 27: "given distance" is very vague here. This is why it would be good to already have a well defined and discussed concept of what the "local region" is and why it is needed in this method.

“Local region” is now defined in the introduction

Page 8, line 7: There is no justification given here for choosing 8 levels.

This is just an illustrative test that shows results under these conditions. (sensitivity to the vertical size of the window is shown in a subsequent figure)

Page 8, lines 11-12: More detail is needed here. Why exactly is this "not a problem"

and "actually an advantage" compared with the direct method?

We have tried to be more explicit: "The default fourth order scheme is slightly non-local, and the direct method would give spurious results very close to the sources; tracking and direct methods would give different results. For example, in the fourth order scheme, if emission are *reduced* in one gridcell, this can reduce the flux from the neighbouring grid cell in the upwind direction, thereby *increasing* the concentration of pollutants in the upwind grid cell.

This is however not a problem for the LF method (or any tracking method), and for short distances it is actually an advantage compared to the direct method."

Figures 5 and 6: Labels are missing for the x-axes. It would also be better to reverse the vertical ordering of the line color legends so that they correspond with the ordering of the lines in the plots.

Good idea, done

Page 11, line 11: Does this mean that more vertical levels would be required when simulating summer months?

Yes, that would be expected. The size of the local region must be adapted to the application. We also added in section 3: "The limitations of the method should be estimated for each concrete application. The examples in this section also provide methods for estimating different errors associated with the method (limitation of the size of the local region, non-linearities)."

Figure 7: Here the size of the local region is referred to by its "distance", whereas everywhere else in the paper the actual size of the local region in grid cells is given. I presume that a distance of 20 is the same as a local region of 41x41, but this is not at all clear. Please use a consistent way of describing the size of the local region.

Yes, done

Page 14, lines 18-19: This seems like speculation (sub-optimal use of cache memory). Another possible explanation is that the extra memory requirements could be leading to increased communication overhead.

It is more than speculation: we had access to more numbers than published. We have rerun these tests, and collected detailed timing information that is now presented in Table 1.

Page 15, line 1: Is this "substantial amount of time" already included in Table 1, or is this additional time? Can the authors quantify this?

The time for writing the data to disk, is (and was) included in Table 1 . We have rerun these tests, and a complete breakdown is now given.

Page 15, lines 2-3: The example given (local region of size 21x21x1) is not used anywhere else in the manuscript. It would be more useful to know about the extra storage requirements of the configurations which are actually evaluated in the manuscript. The authors should expand Table 1 to also include the extra storage requirements of the configurations given in this Table.

Done

"for instance, for a local region of size 81x81x10 and for each sector or species, $400 \times 260 \times 81 \times 81$ values = 2.5 GB of data have to be written to disk each time it is requested (only one vertical level is written out). The corresponding memory demand is calculated in the same way, but must further be multiplied by the number of vertical levels of the local region, then by two because one array is needed to store the instantaneous values and one for accumulating the values over time, and multiplied by another factor two because the calculations are done in double precision."

Page 15, line 5: "origin of pollutants" should acknowledge the limitations of the method.

Based on the evaluation presented by the authors, it seems that this method can currently only analyse the origin of some kinds of primary pollutants.

Added the word "primary"

Page 15, lines 11-13: The authors have not provided any other details about interactive graphical user interfaces. Is this something for future work? Or can the authors already provide a reference for this?

We have a python script that works as a demo version. This is not meant to be the first version to be developed, but rather something basic that helps to look at the Local Fractions, as it is many dimensional and standard tools are not always adapted. We could put it on zenodo, together with a sample of data. Here is a temporary link (<https://sandbox.zenodo.org/record/455127>). If you recommend it, we can put it as permanent and reference it in the paper.

We have plans to make a more useful LF-GUI, but this has not concretized yet.

Page 15, line 17: This is an important point, and not necessarily a disadvantage of the method. For some applications it may be acceptable to simply know that a certain amount of pollution originates from outside the local region. This provides some justification for other trade-offs which are made when using this method.

Page 15, line 19: Can the authors go into more detail about the "double counting" problem and how their approach solves it?

Page 15, lines 23-24: Which of the "several" problems are avoided and how? This text is way too vague.

This section has been revised. Also added "In a city, scales down to street level may be required. Those very fine scale models will not compute accurately the transport between distant streets within the city and the regional model must account for those. But if the same emissions source are included both in the regional and fine scale model they will be accounted for twice."

Page 16, line 14: It seems to me that the local fractions deliver information about contributions, not sensitivities.

Page 16, line 14: Why wouldn't the local fractions add up to 100%, and why isn't this a problem? It seems that the final sentence of the manuscript creates all sorts of problems for the interested reader. The authors could consider simply deleting this sentence, and merging the previous sentence with the previous paragraph.

We really meant to say sensitivities. But we can agree that without further details, it does not give enough explanation. So rather than explaining the details of what we might do, we have deleted this sentence and replaced it by

"In the future we plan to generalize the method to also include chemical processes in some simplified form. The ambition is to still provide information for a very large number of sources, but to describe chemical processes in an approximate way. Compared to existing tagging methods, it will trade accuracy for computational efficiency."

Local Fractions - a method for the calculation of local source contributions to air pollution, illustrated by examples using the EMEP MSC-W model (rv4_33)

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Abstract. We present a computationally inexpensive method for individually quantifying the contributions from different sources to local air pollution. It can explicitly distinguish between regional/background and local/urban air pollution, allowing fully consistent downscaling schemes.

The method can be implemented in existing Eulerian chemical transport models and can be used to distinguish the contribution of a large number of emission sources to air pollution in every receptor grid cell within one single model simulation and thus to provide detailed maps of the origin of the pollutants. Hence it can be used for time-critical operational services providing scientific information as input to local policy decisions on air pollution abatement. The main limitation in its current version is that non-linear chemical processes are not accounted for and only primary pollutants can be addressed.

In this paper we provide a technical description of the method and discuss various applications for scientific and policy purposes.

1 Introduction

The origin of atmospheric pollutants within a given region is one of the fundamental questions of air quality research. Degradation of air quality, either temporary or sustained, is often the result of both local and long-range transported air pollution, originating from anthropogenic but also natural emission sources. Anthropogenic emissions are due to a large number of different categories such as road traffic, industrial point sources and large area sources.

In order to devise optimal strategies of air pollution abatement, for example short-term or long-term emission reduction measures, air quality managers need to have access to reliable scientific knowledge about the origin of air pollution. Typical questions include: a) By what amount can local air pollution be reduced through local measures only, and in which cases will regional or countrywide measures be necessary? b) What will be the benefit of emission reduction measures imposed on one or several specific emission sectors? c) Will these measures be efficient on a short time frame or should they be implemented on a longer-term basis?

Many different methods exist to extract information about the origin of air pollution (e.g., Thunis et al., 2019; Clappier et al., 2017). Some of them are based on measurements of the chemical composition of air masses in the region or interest (receptor region). Such a ‘chemical fingerprint’ can then give hints on the origin (source region or sector) of the pollutants. Most methods, however, are based on models as these can be readily applied to scenario calculations as well. Chemical Transport Models (CTMs), in particular, are efficient mathematical tools that treat emission sources, transport, chemical conversion and loss mechanisms of air pollutants in a consistent way, and allow different scenarios to be assessed within a reasonable amount of computing time.

The simplest method to evaluate the importance of different emission sources in a CTM is the ‘direct’ method (e.g., Folberth et al., 2012), sometimes also referred to as ‘annihilation’ or ‘brute force’ method, where the same model simulation is repeated with and without including a chosen emission source. The difference in pollutant concentrations in the receptor region can then be attributed to the chosen emission source (impact). In order to stay within quasi-linearity one can choose to reduce the emission source by only a small amount. This is usually referred to as ‘perturbation method’ (e.g., Jacob et al., 1999; Fiore et al., 2009) and is well suited to simulate the effects of policy measures to reduce emissions from certain sectors by a certain amount. However, one of the drawbacks of this method is that for each source contribution a new, independent, simulation must be performed.

In general, the chemical processes involve non-linearities, and the sum of individual ~~contributions–differences~~, calculated by the direct or perturbation methods does not equal the base case simulation where all sources are included. ~~A method that better treats non-linearities is~~ For Ozone and other non-linear species, it is crucial to distinguish between the effect of a change in emissions (sensitivity) and the relative contribution of different sources (source apportionment). An alternative approach for determining source–receptor relationships in model runs is a technique known as “tagging”, in which ozone molecules are labelled with the identity of their source, allowing direct attribution of ozone concentrations to these sources in receptor regions. A series of methods have been proposed to address calculate directly the contribution of different sources, based on the so-called ‘tagging method’ (e.g., Wang et al., 1998; Wu et al., 2011; ? (e.g., Butler et al., 2018; Emmons et al., 2012; Dunker et al., 2002; Kwok et al., 2002). It distinguishes chemically identical molecules according to their sources. In the model this can be done by defining one separate tracer for each source. In this way it is possible to track these tracers through transport and chemical transformations, and thereby quantify their contribution to air pollution at any given location in one single model run. ~~The limitation of tagging methods has been that~~

Tagging methods are also useful for tracing primary pollutants (e.g., Kranenburg et al., 2013). However, in cases where the number of different tagged sources is large, the calculation becomes unpractical, the tagging methods can become excessively computationally expensive.

In this regard, ‘adjoint models’ (Elbern and Schmidt, 1999; Vautard et al., 2000; Henze et al., 2007) are superior. Adjoint models calculate the derivative of a model scalar with respect to all other model parameters in one single simulation and in this way efficiently quantify the contribution from all emission sources to air pollution in a given receptor region. However, a new adjoint simulation must be performed for each receptor region.

Still, only a relatively small amount of sources or receptors at a time can be analyzed by all these methods. Perturbation methods calculate all receptor values for one source group, tagging methods compute all receptors for a limited number of source groups, while adjoint models address all sources for one receptor group. Ideally, all contributions to all receptor points should be described.

5 In this paper we present a method which can efficiently calculate the contribution of a significantly larger amount of sources (thousands or more), to a limited (but large) number of receptor regions. This method does not provide results that cannot be obtained by other means, but it does so at a lower computational cost and is thus well suited especially for time critical operational applications. It can be built on top of existing Eulerian CTMs relatively easily, and thereby has the potential to offer a new range of applications. ~~However, it is at present~~

10 ~~An important limitation is that the method is~~ limited to primary pollutants, for which linearity can be assumed. It will thus complement existing methods, but not replace them.

~~In principle the method allows the definition of any group of sources, but here we will show results only for the case where each defined source is defined within a single grid cell. One key limitation, which makes the method manageable, is that the tagged values are stored only up to a preset horizontal and vertical distance from their source. We will call the region within~~
15 ~~this distance the "local region". The size of this region must be set as a balance between computational cost and the accuracy requirement of the application.~~

~~In the following Section we describe the method in technical detail, while in Sect. 3 we show concrete examples of what kind of results the method can provide, and how to quantify some of the limitations associated with the method. The results will also be compared against the direct method. In Sect. 4 we will give an overview over what is required to implement the~~
20 ~~method in an existing CTM and discuss the performance in the EMEP MSC-W implementation.~~ Finally in the last section we discuss possible applications of the method as well as plans for further development.

2 Description of the method

In theory the method corresponds to a tagging method, where pollutants from different origins are tagged and their values are traced and stored individually. ~~However the total amount of pollutants is not computed as a sum of tagged values; instead the~~
25 ~~tagged values show which fraction of the total pollutants originate from a specific origin.~~

We define the Local Fraction LF_s in a receptor grid cell as the fraction of pollutant that is due to a particular source term S . For example, S can be a power plant or the road traffic emissions in a specific source region. LF_s is a number between zero and one and is calculated as:

$$LF_s = \frac{\text{Pollutant due to source } S}{\text{Total Pollutant}} = \frac{LP_s}{TP} \quad (1)$$

30 The Total Pollutant is abbreviated TP ; it could be the air concentration of particulate matter for example. The Local Pollutant, LP_s , is the part of TP "tagged" from a specific origin S . Its value is in general the result of various processes (emissions,

advection, diffusion, etc.) as will be described below. Given the value of the Total Pollutant, then the Local Fraction and the Local Pollutant carry the same information, but as we will see, there are a few practical advantages of storing the Local Fraction rather than the Local Pollutant.

In a time-splitting framework the different physical processes are included sequentially, and we will show in the next Sections how the value of the Local Pollutant changes during each of them. For simplicity, the initial value for LF_s is set to zero, given that in the long term LF_s should not be sensitive to the initial value.

2.1 Emissions

The Local Pollutant and Local Fraction are associated with a particular emission source category (E_s) in a specific grid cell. (Formally the source could also be spread over a group of grid cells, but at present we limit ourselves to sources defined on single grid cells). If $E_s(t)$ is the emission rate of source s at time t , LP_s will increase during the time step Δt :

$$LP_s(t + \Delta t) = LP_s(t) + E_s(t)\Delta t \quad (2)$$

and

$$LF_s(t + \Delta t) = \frac{LP_s(t + \Delta t)}{TP(t + \Delta t)} \quad (3)$$

For instance s could refer to emissions of particulate matter from road traffic emissions TP would be the total concentration of particulate matter in the receptor region, and LF_s would then be the fraction by which the total concentrations in the receptor region would be reduced if the emissions from road traffic in the source region were removed completely (assuming linearity).

2.2 Advection

Transport of pollutants will mix pollutants from different origins. We will trace individually the Local Pollutant due to different sources and from every horizontal grid cell within the source region. We need then two sets of position indices, one for the origin (source region) and one for the actual position (receptor grid cell):

$$LF_{s,x_s,y_s}(x, y, z, t) \quad (4)$$

Where x_s and y_s are the (horizontal) coordinates of the source grid cell, and x, y and z are the coordinates of the receptor grid cell. s is a specific source category at (x_s, y_s) . In order to keep the calculation at a reasonable cost, one can limit x_s and y_s to be within a preset number of grid cells from the receptor grid cell, Δ^{max} :

$$x - \Delta^{max} < x_s < x + \Delta^{max} \quad y - \Delta^{max} < y_s < y + \Delta^{max} \quad (5)$$

The source position indices are then replaced by its relative position relative to the receptor grid cell:

$$LF_{s,\Delta x_s,\Delta y_s}(x, y, z, t) \quad (6)$$

where $\Delta x_s = x_s - x$ and $\Delta y_s = y_s - y$ are the signed distances to the source. In practice, also z is limited, as it is usually not necessary to trace pollutants for receptor grid cells all the way up through the atmosphere. Note that the vertical position of the source is not explicitly traced, but it can, in principle, be included in the form of separate sources S .

We call the region delimited by all (x_s, y_s) and the vertical range of z for the "local region".

5 $LF_{S, \Delta x_s, \Delta y_s}(x, y, z, t)$ in practice a seven dimensional array. The range of S depends on the number of source categories to be tracked. The size of this array can be very large, which reflects the large amount of information it carries.

Pollutants can be traced within this region. If they leave the local region, they are no longer identifiable by the method, even if they return into the local region.

10 Let us consider a flux of pollutant, $F(x, y, z, t)$ (assumed positive), from a grid cell x to $x + 1$ during Δt , and a source at a position Δx_s relative to x .

The amount of Local Pollutant leaving the grid cell x is

$$F(x, y, z, t) LF_{S, \Delta x_s, \Delta y_s}(x, y, z, t) \quad (7)$$

At position $x + 1$ the relative position of that source is $x_s - 1$ and the Local Pollutant is thus updated according to

$$LP_{S, \Delta x_s - 1, \Delta y_s}(x + 1, y, z, t + \Delta t) = LP_{S, \Delta x_s - 1, \Delta y_s}(x + 1, y, z, t) + F(x, y, z, t) LF_{S, \Delta x_s - 1, \Delta y_s}(x, y, z, t) \quad (8)$$

15 Or, if the source is moved by one grid cell (Δx_s replaced by $\Delta x_s + 1$), the formula becomes:

$$LP_{S, \Delta x_s, \Delta y_s}(x + 1, y, z, t + \Delta t) = LP_{S, \Delta x_s, \Delta y_s}(x + 1, y, z, t) + F(x, y, z, t) LF_{S, \Delta x_s + 1, \Delta y_s}(x, y, z, t) \quad (9)$$

The Local Fractions are then updated according to the definition in Eq. (1).

$$LF_{S, \Delta x_s + 1, \Delta y_s}(x + 1, y, z, t + \Delta t) = \frac{LP_{S, \Delta x_s + 1, \Delta y_s}(x + 1, y, z, t + \Delta t)}{TP(x + 1, y, z, t + \Delta t)} \quad (10)$$

20 The fluxes and Total Pollutants are not explicitly dependent on the source S , and are normally available quantities in the CTM model.

If the flux is exiting the grid cell x , the Local Fractions at x do not have to be updated, since it can be assumed that the fractions being removed are the same for the Local and Total Pollutants.

2.3 Diffusion (and convection)

For diffusion we compute the effect of diffusion directly on every Local Pollutant:

$$25 LF_{S, \Delta x_s, \Delta y_s}(x, y, :, t + \Delta t) = \frac{\text{Diffusion}(LP_{S, \Delta x_s, \Delta y_s}(x, y, :, t))}{\text{Diffusion}(TP(x, y, :, t))} \quad (11)$$

Where “Diffusion()” is the numerical operator that computes the diffusion in the model and the colon ‘:’ indicates its operation over the entire vertical grid column. This ensures a consistent treatment of the Local Fractions, whatever numerical procedure is applied for the diffusion.

In a practical implementation it is not necessary to include all the vertical levels, as the contribution from higher levels is negligible (it corresponds to pollutants leaving and returning to the local region during the same time step). In our implementation we include only two layers above the highest local region considered.

For convection the same procedure can be used by replacing the diffusion operator in Eq. (11) by the convection operator. In the current EMEP MSC-W model version, the convective processes are not implemented in the Local Fraction calculations.

10 2.4 Deposition

For deposition (dry or wet), we can assume that the same fractions of Local and Total Pollutants are removed. Therefore the Local Fraction will not vary during the deposition process:

$$LF(t + \Delta t) = LF(t) \quad (12)$$

The simplicity of this formula is one of the motivations for storing LF rather than LP .

15 2.5 Chemistry

To fully follow the pollutants through all the chemical reactions would, in principle, require an explicit reference to all the sources and grids. It is possible to reduce the size of the problem if linearity is assumed. This has been done by other groups (e.g., Kranenburg et al., 2013). The calculation of all the chemical reactions is one of the most computationally intensive part of CTMs (roughly 60% in the EMEP MSC-W model (Simpson et al., 2012) used for the tests presented below). A consistent chemical treatment of Local Pollutants would mean to almost multiply the computation time by the number of Local Pollutants considered, i.e. the size of $(S, \Delta x_s, \Delta y_s)$. In order to preserve the simplicity of the method, we will in this version assume that the chemical processes modify the local and non-local part of the pollutants in the same proportions. With this assumption Eq. (12) can be used. This assumption is correct for primary particles and, as illustrated in our examples below, can give meaningful results also for NH_3 , SO_x and NO_x . So far the method is only developed for emitted pollutants, and not for secondary pollutants.

3 Examples and validation

The Local Fractions will depend on a broad range of factors such as emission distributions, meteorological conditions, grid resolution, chemical regime, size of the local region etc. It is beyond the scope of this article to systematically quantify how all the possible situations affect Local Fractions. ~~Here we will only try to give some basic examples for situations where the approximations made are valid and the results calculated are of relevance.~~ The limitations of the method should be estimated

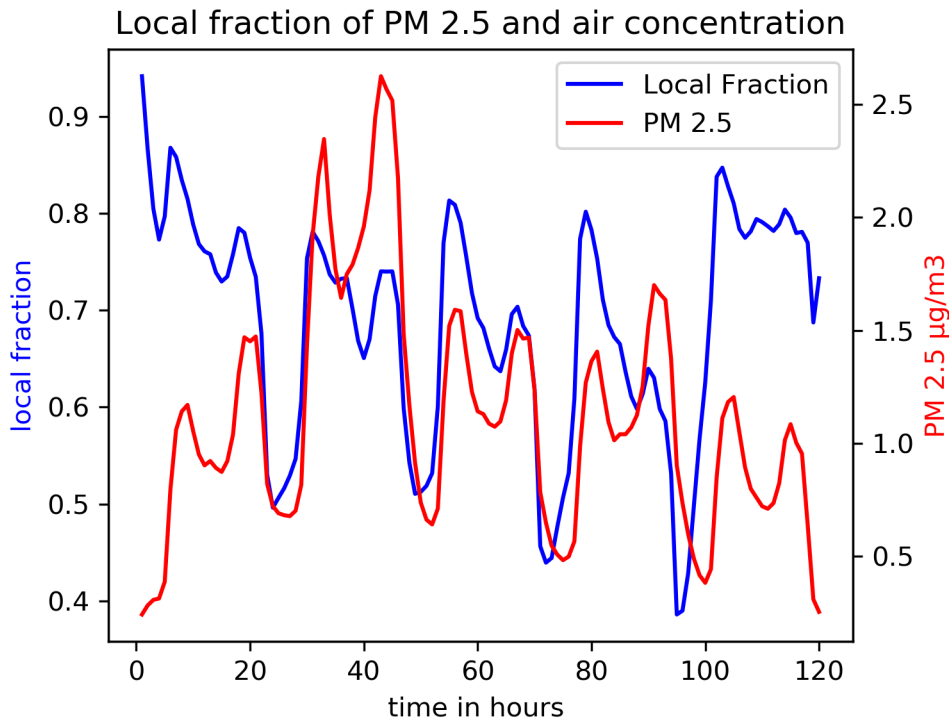


Figure 1. Time evolution of the Local Fraction of PM_{2.5} in the Oslo agglomeration (left axis) during the period 5th to 9th January 2016 (longitude=11.55°, latitude=59.9°). The total concentration of PM is also shown (right axis).

[for each concrete application. The examples in this section also provide methods for estimating different errors associated with the method \(limitation of the size of the local region, non-linearities\).](#)

The Local Fraction $LF_{s,\Delta x_s,\Delta y_s}(x, y, z, t)$ is a 7-dimensional array, and in the following Sections we will try to briefly illustrate the information that can be provided by this array.

- 5 The results shown in this Section are based on a grid with a resolution of 0.3° in the longitude direction and 0.2° in the latitude direction. [The parameter settings are essentially the same as what is used for the official EMEP MSC-W model runs, using "TNO MACC-III" emissions \(2015 update of \(Kuenen et al., 2014\)\). However to simplify the interpretation of the results, two important modifications have been introduced: a simplified advection scheme is used \(see Sec. 3.2\), and all emissions are released at the lowest level. The standard settings of the model do not include convection over Europe.](#)

10 3.1 Time and space dependence ($\Delta x_s = \Delta y_s = 0$)

In Fig. 1, an illustration of the time evolution of the instantaneous Local Fraction for fine particulate matter (PM_{2.5}) at an arbitrary location (in the Oslo agglomeration) is shown. The value gives the fraction of PM_{2.5} which has its origin in the same grid cell. It is strongly correlated with the concentrations of PM_{2.5}, but it does not always vary exactly in the same way. It will

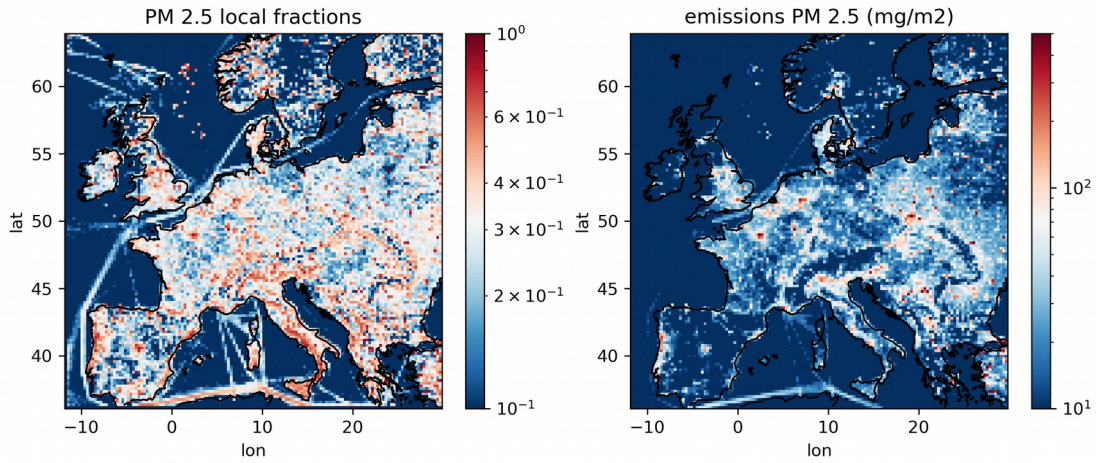


Figure 2. Example of spatial distribution of the Local Fraction of PM_{2.5}, averaged over one month (March 2016, left panel). The total emissions of PM_{2.5} averaged accumulated over that period are shown in the right panel.

also depend on the wind speed, emission rates and the surrounding levels of pollution. If a relatively large amount of clean air is moving into that area, the total concentration will decrease, but the Local Fraction will remain high. High Local Fractions indicate that most of the pollutant is locally produced.

Figure 2 shows a map of monthly-mean Local Fractions for March 2016. It gives a picture of how much the sources in a particular grid cell contribute compared to the surrounding sources. The distribution is similar to the emission distribution, but isolated emission sources show up more clearly in the Local Fractions map.

20 3.2 Illustration of Source Receptor capabilities

For a fixed value of x , y and z and t , the Local Fractions $LF_{s,\Delta x_s,\Delta y_s}(x, y, z, t)$ give the contributions of a pollutant S emitted at $(x + \Delta x_s, y + \Delta y_s)$ to the position (x, y) i.e. a two dimensional map of the origin of the pollutants found at position (x, y) . Thus provides a complete description of all source receptor relationships within a given distance from the receptor grid cell.

Figure 3 shows such a map for an arbitrary location. It is simply the value of $LF_{s,\Delta x_s,\Delta y_s}(x, y, z, t)$ averaged over one month, where x and y are the position of the central point (receptor). Such a map is calculated for any point on the grid in a single simulation. In this example the local region has a horizontal extend of 41 times 41 grid cells. Direct methods would then, in principle, require $41 \cdot 41 + 1 = 1682$ calculations to calculate the values of one of those maps.

5 In order to compare with the direct method, one can "invert" $LP_{s,\Delta x_s,\Delta y_s}(x, y, z, t)$ to get a map of the receptors for a fixed source:

$$LP_{s,\Delta x_s,\Delta y_s}^\dagger(x, y, z, t) = LP_{s,-\Delta x_s,-\Delta y_s}(x + \Delta x_s, y + \Delta y_s, z, t) \quad (13)$$

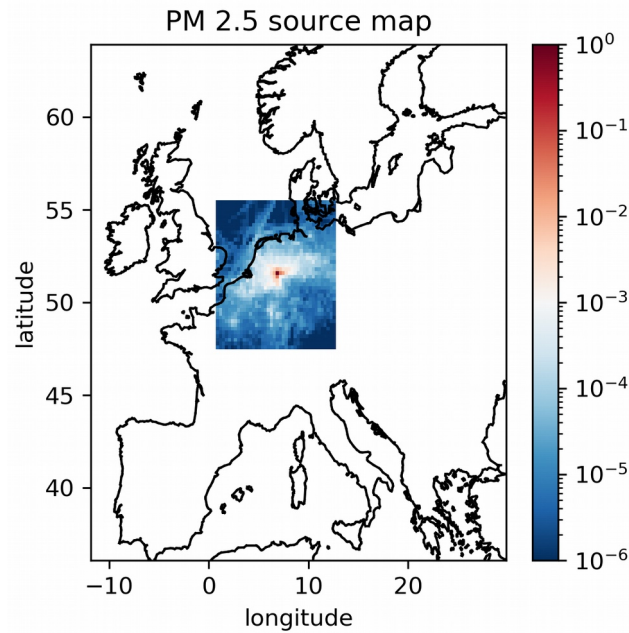


Figure 3. Example of Local Fractions as a source map. The values show the fraction of $PM_{2.5}$ that has been emitted at that location and transported to the central point. The sum of all the fractions is in this case 0.976, meaning that 2.4 % of the $PM_{2.5}$ concentration at the central position, originates from sources outside of the local region

$LP_{s, \Delta x_s, \Delta y_s}^t(x, y, z, t)$ then gives the contributions of a pollutant s located at (x, y) to the position $(x + \Delta x_s, y + \Delta y_s)$

Figure 4 illustrates a comparison of the results obtained

- 1) by removing the emissions from a single grid cell and computing the difference with the normal case (direct method).
- 2) by using one single run and Eq. (13) with a local region of size $41 \times 41 \times 8$.

Within the local region the results are similar, but the Local Fraction method gives such a map for any grid cell in one single run, while the direct method would require a separate run for each source region.

Note that for the purposes of this experiment we have chosen a zero order advection scheme in all model runs. The default fourth order scheme is slightly non-local, and the direct method would give spurious results very close to the sources tracking and direct methods would give different results. For example, in the fourth order scheme, if emission are reduced in one gridcell, this can reduce the flux from the neighbouring grid cell in the upwind direction, thereby increasing the concentration of pollutants in the upwind grid cell. This is however not a problem for the LF method (or any tracking method), and for short distances it is actually an advantage compared to the direct method.

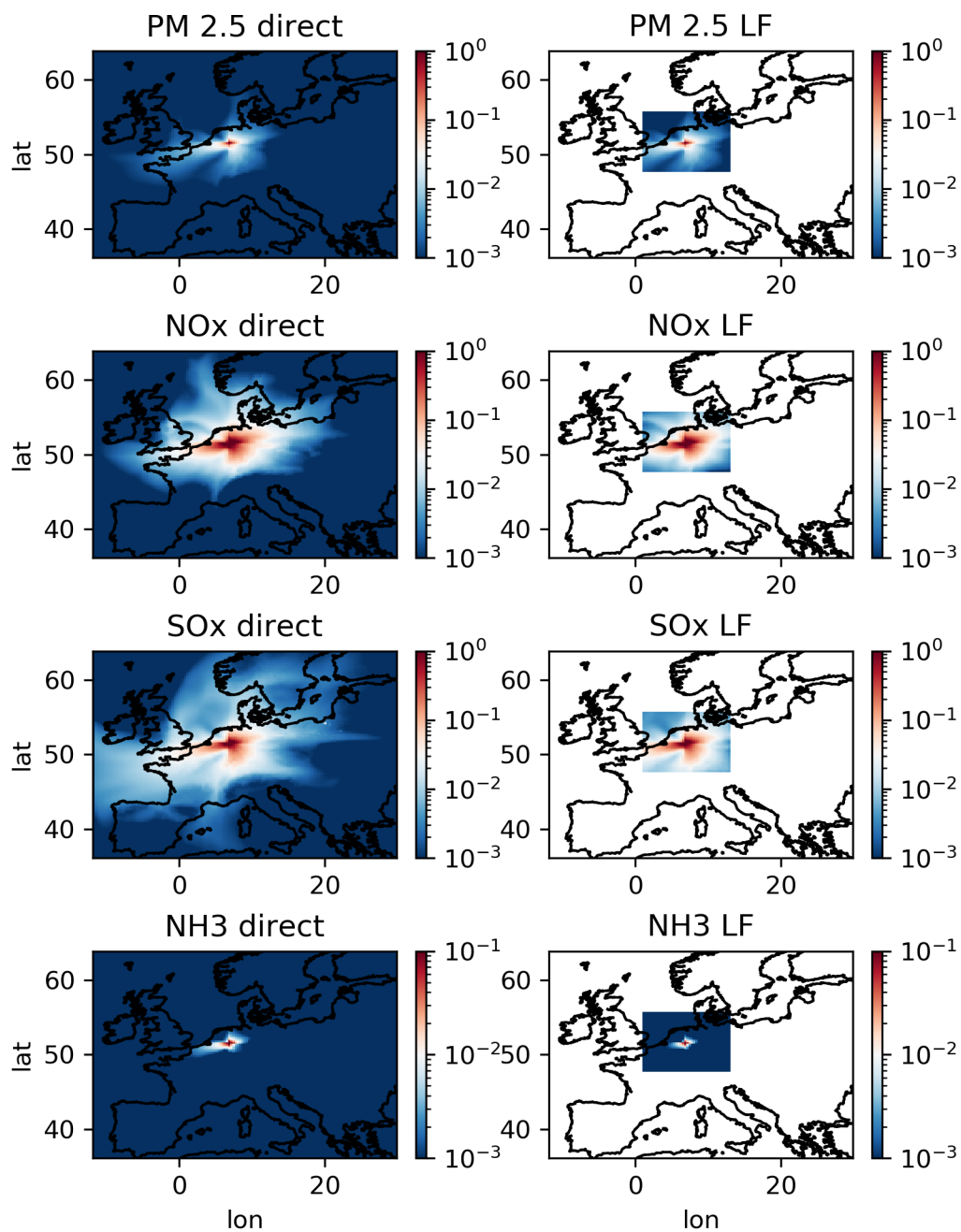


Figure 4. Receptor map for a single grid cell emission, obtained through direct method (left panels) and the Local Fraction method (right panels), averaged over one month (March 2016). Concentrations of PM_{2.5}, NO_x, SO_x and NH₃ (in μg m⁻³). The direct method requires a separate run for each source location. The Local Fraction method gives the receptor map in one single run, in a limited region around the source, but for any source grid cell.

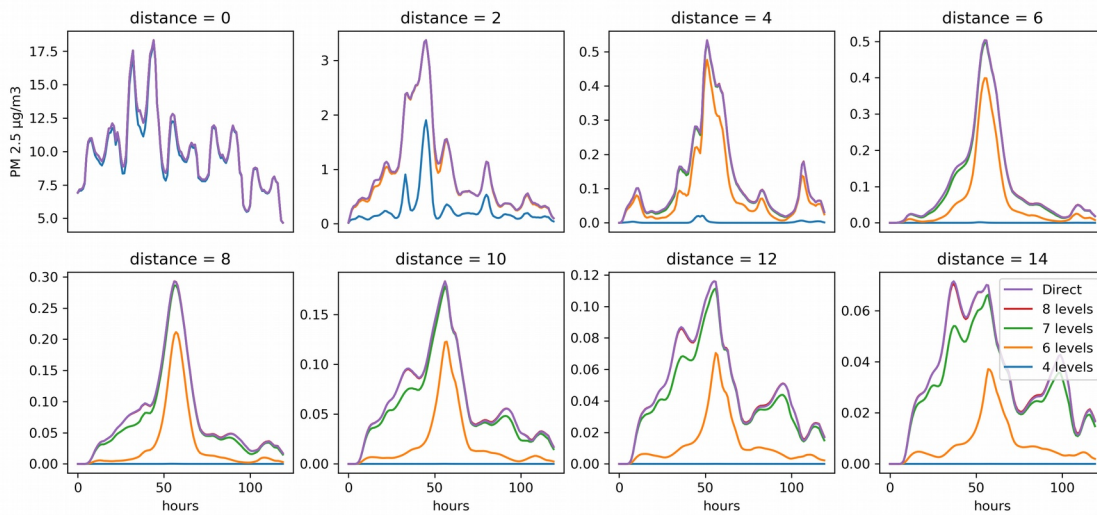


Figure 5. Sensitivity of the concentration of PM_{2.5} ($\mu\text{g m}^{-3}$) to the number of vertical levels included in the local region, for different distances from the source. The distance from the source is given in numbers of grid cells (one grid cell = 0.3 degrees in longitude direction). Source in Oslo agglomeration. Horizontal axis is time (120 hours). The 8 levels results cannot be distinguished from the direct method on the figure, even for the largest distance considered.

3.3 Vertical transport

For source apportionment applications, the focus is typically on horizontal transport. Nevertheless the code should trace the pollutants with a combination of vertical and horizontal transport. Over short distances only transport through the lowest layers needs to be considered. If the focus is on regions where a large part of the pollutants are transported over long distances, the vertical extend of the local area should be chosen large enough.

Figure 5 illustrates the dependence of the Local Fraction on the thickness of the local region. In this example, only a few vertical levels are required to describe the Local Fraction within the grid cell (the remaining discrepancy comes from pollutants first leaving the grid cell, and then returning later). For a distance of up to 14 grid cells, including 8 vertical layers in the local region, results are not distinguishable from the exact value calculated by the direct method. Obviously, emission or vertical mixing at higher altitudes would require to include the corresponding vertical layers.

For NO_x, even for relatively small distances, there is a discrepancy between the contribution calculated with the Local Fraction method and the direct method (Fig. 6). This is because the Local Fraction method does not explicitly distinguish between NO and NO₂. The mix modelled in the remote emissions may differ from the local values. Since reaction rates are different for NO and NO₂, the local NO_x transformation rate is not representative for the reaction rates of the incoming "older" NO_x.

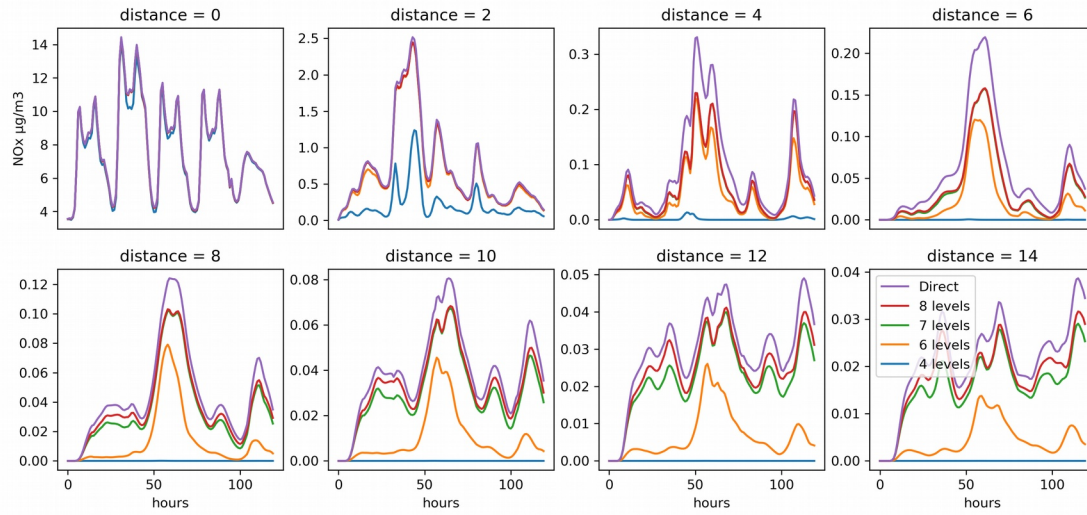


Figure 6. Concentration of NOx ($\mu\text{g m}^{-3}$). Sensibility to number of vertical levels included in the local region. Distance to source in number of grid cells (one grid cell = 0.3 degrees in longitude direction). Source in Oslo agglomeration. Horizontal axis is time (120 hours). At a distances larger than a few grid cells, a discrepancy can be observed between the contribution calculated with the Local Fraction method and the direct method.

3.4 Completeness

For local regions that are large enough, the source of all primary particles can be accounted for. This can be verified directly by summing all the Local Fractions for a given grid cell:

$$\sum_{\Delta x_s, \Delta y_s} LF_{s, \Delta x_s, \Delta y_s}(x, y) \quad (14)$$

- 5 A sum of one means that all sources are accounted for. The difference between the sum of of the Local Fractions and one gives the fraction of pollutants with sources outside of the local region. In Fig. 7 the sum of the Local Fractions is shown for every grid cell on the map for different horizontal sizes of the local region. For most land areas, more than 80% of the sources are found for the smallest window (41x41) and essentially all sources for the largest (161x161).

10 Figure 8 show the result for different vertical extend of the local region. The Local Fractions get close to complete in most places, when 8 vertical levels are included (approximately 1522 meters height). As one would expect, this roughly corresponds to the maximum height of the boundary layer in March over land in Europe.

Note that incomplete results are not a measure of an error in the method. Rather they show the amount of pollutants with sources outside of the local region, which is useful information.

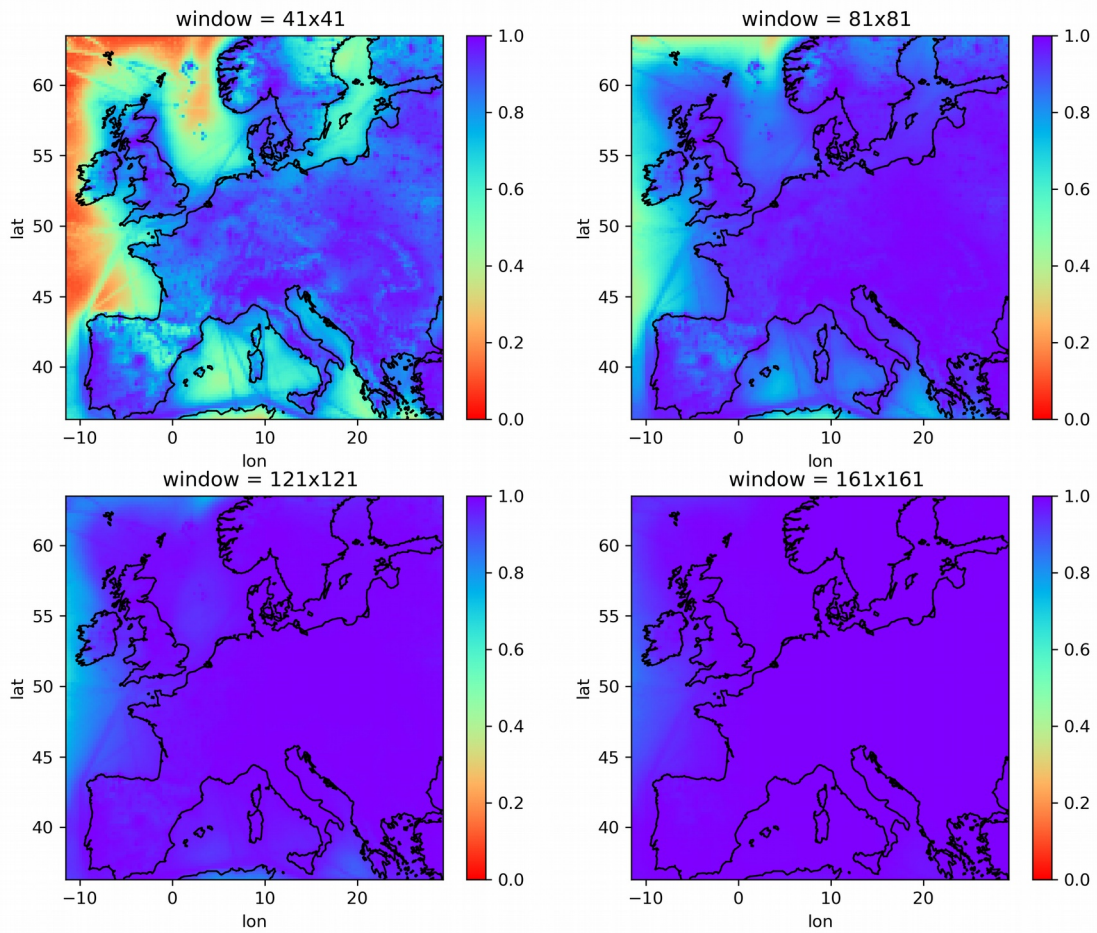


Figure 7. Sum of all Local Fractions (Eq. (14)) for PM_{2.5} and different sizes of the local region (average for March 2016). The distance is counted as number of grid cells in each direction. All vertical layers (20) are included. A sum of 1.0 means that all the sources have been accounted for.

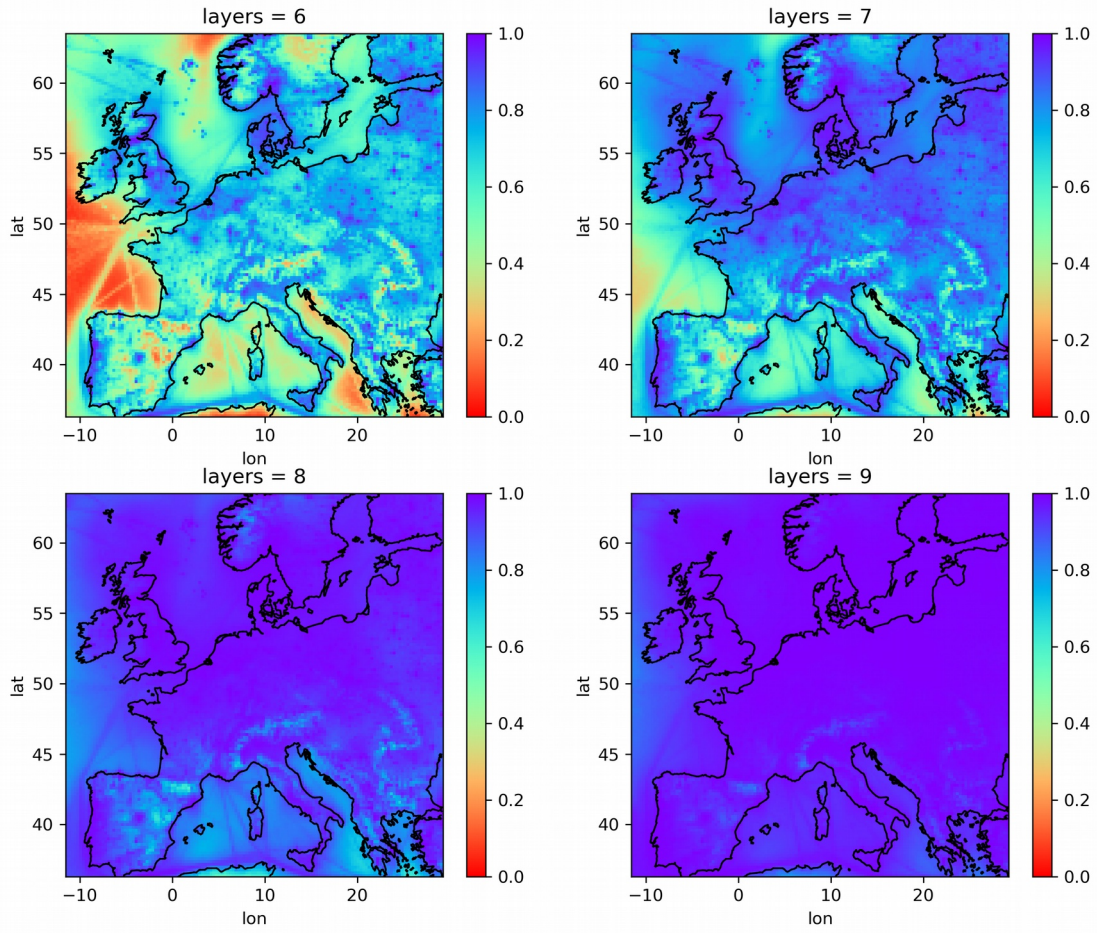


Figure 8. Sum of all Local Fractions (Eq. (14)) for PM_{2.5} and for different vertical extents of the local region (average for March 2016). A horizontal region of 161×161 is included in the local region. For a standard atmosphere, the height of the top of the layers 6, 7, 8 and 9 are, respectively, 623, 1015, 1522 and 2149 meters. A sum of 1.0 means that all the sources have been accounted for.

3.5 Computational aspects

15 4 Implementation and Computational aspects

From an implementation point of view the method is a "diagnostic" calculation, in the sense that it gives additional information extracted from existing data, in opposition to a modification of the method for computation of the concentrations of air pollutants. Therefore the method can be implemented on top of existing CTM, without having to rewrite the code for the main processes. What is required is to include calls to new routines that can perform the operations described in Sect. 2.

20 Concretely, the main changes to be made are:

- Define the instantaneous Local Fraction 6-dimensional array $LF(S, \Delta x_s, \Delta y_s, x, y, z)$ and one corresponding array for each of the time averaged periods (at least one for averaging over the run, and possibly another for averaging over hours for example).
- Write a routine that performs the operations from Sect. 2. In addition a routine for writing out the results (i.e. six dimensional Local Fraction arrays) and one routine should do the averaging over time.
- As input for those routine, the main code must make available the emission rates of the relevant sectors and the advection fluxes. If the fluxes are not available, or in an early version, the fluxes could be defined directly by a simpler method. For example an already good approximation would be to take $F_x = \frac{c\Delta t}{\Delta x} LP_{up}$, where c is the wind speed in x direction, Δx the size of the gridcell and LP_{up} the concentrations in the upwind gridcell.
- In Eq. 9 it is necessary to have access to values from the nearest neighbour gridcells. In a parallel implementations, this may require supplementary communication routines.
- In addition, of course, the calls to those new routines have to be integrated into the main code. Also switches to choose the pollutants and the sizes of the local region have to be created.

5 There is no feedback of the LF calculations to the concentrations of air pollutants; those will be unaffected by the new routines. This clear separation greatly simplifies the practical development.

In the EMEP MSC-W implementation (rv4_33), all the extra routines are put in a separate file ("uEMEP_mod.f90"), except for the LF communication routine. If no LF output is required, those routine are not used at all, and if the LF routine are called, the rest of the code still performs exactly the same operations.

10 Since one of the key advantages of the Local Fraction method is its low computational demand, we will give a few concrete examples of the computational cost for providing the Local Fraction values in our implementation. The transformations carried out for the calculation of Local Fractions presented in Sect. 2 are all relatively simple. The most computationally intensive parts of the model (calculation of fluxes, chemical transformations, deposition processes) are not explicitly performed for every Local Pollutant, but only once for the total concentrations. ~~This also means that there are no fundamental changes in the model code involved. The updates of the Local Fractions can be added on top of an existing model in separate routines.~~ — For processes

window-size	local region	levels	total time	additional time	emission	advection	diffusion	write	averaging	comm.	other	mem
11x11	11x11x3	3	1.9 %	1.6%	0.5	0.8	0.2	0.4	0.1	0.3	-0.4	0.6
21x21	21x21x6	6	11.9	11%	0.6	5.3	1.3	1.2	0.5	2.4	0.7	4.0
51x51	41x41x6	6	38.5	39%	0.9	18.4	4.5	3.8	1.6	9.4	-0.0	16.0
51x51	41x41x10	10	63.0	64%	0.9	31.6	6.9	3.8	2.9	17.1	-0.3	26.0
81x81	81x81x10	10	241.3	240%	2.0	117.5	27.9	14.3	13.9	67.3	-1.6	102.0
121x121	121x121x10	10	623.7	621%	4.7	292.2	108.3	30.2	32.8	170.5	-15.0	227.0
161x161	161x161x10	10	1472.1	1610%	11.3	740.3	284.4	51.6	62.4	336.1	-14.1	402.0

Table 1. Additional computation time needed for the calculation of Local Fractions in different settings, expressed as fraction of in % in comparison to the total time needed when calculation of Local Fractions is not included. The first column shows the horizontal-size dimensions of the local region, while the second column shows the total additional time required. Column three to eight show the breakdown of those fractions in the different subroutines ("comm." stands for communication time between compute nodes). "other" show the difference between total time and its components (it is principally due to uncontrolled differences in speed of the different compute nodes). The last column shows the additional memory required in total. It has to be multiplied by the number of vertical levels through which pollutants are traced species or sectors requested. The total time without calculation of Local Fractions in our tests was 447.553 seconds.

were local pollutants are transformed by the same relative amount as non-local pollutants (deposition and chemistry in our implementation), there is no need to update the Local Fractions; this is the main motivation for storing the Local Fractions rather than the local pollutants.

The calculation of the Local Fractions only needs information from the nearest neighbors, see Eq. (9) and is therefore well suited for parallel processing in a space partition framework. While storing all the Local Fractions is memory demanding, the data are distributed among the compute nodes, so that the memory requirement can be met by increasing the number of nodes.

In order to illustrate the computational cost, we can consider a typical model run, on a $400 \times 260 \times 20$ grid (0.3 degrees longitude \times 0.2 degrees latitude resolution), over one month (March 2016) on 160 processors that takes 447—553 seconds without the Local Fraction calculations. Table 1 shows the additional computational cost for computing the Local Fractions in our implementation. The mathematical operations required to compute the Local Fractions are proportional to the number of sources considered and the size of the local area (in our implementation the additional time required for those operations advection and diffusion grows faster, probably because of sub-optimal utilization of cache memory). If one is only interested in the nearby sources (within a city, for example), the Local Fractions can be calculated at almost no additional cost. Remote sources can still be described, but at an additional cost.

A substantial amount of time can be required for writing results to disk, specially if all results are written out every hour—required at finer time resolution, for example every hour (in Table 1 the results are only written out once). This is mainly due to the large amount of data collected; for instance, for a local region of size $81 \times 21 \times 21$ and 14 sectors, 10 and for each sector or species, $400 \times 260 \times 21 \times 21 \times 14 \times 10$ values = 5.25 GB of data have to be written to disk each time it is requested (only one vertical level is written out). The corresponding memory demand is calculated in the same way, but

must further be multiplied by the number of vertical levels of the local region, then by two because one array is needed to store the instantaneous values and one for accumulating the values over time, and multiplied by another factor two because the calculations are done in double precision.

5 Discussion

- 5 Local Fractions are a new concept that can help understand and analyse the origin of primary pollutants. It has the potential to be developed further, and a new range of applications is still being developed.

Compared to other approaches, there are always trade offs. The present method cannot at present describe non-linearities. That excludes all study of Ozone. Long range transport will also become unpractical at some point, although this is not inherent to the method and could be implemented in the near future.

10 5.1 Source apportionment

Source receptor relationships can be produced for any source and receptor within a region around the source. The size of this region can be chosen to be relatively large (100 grid cells or more). Since the fluxes are given from and to individual grid cells, small regions (typically cities) can be studied simply by adding up individual grid cell contributions. These small regions do not have to be predefined in the model simulations. Indeed, the relative contributions of sources that contribute to the pollutants within a city covering several grid cells can be determined in a post-processing step, using graphical user interfaces where the user can choose the source region and source categories interactively.

Still, the method provides information about transport within a limited region only (the 'local region'). The choice of the size of this region is a balance between the computational cost and the distance to the sources of interest. For the study of a city, it may be sufficient to include a region covering the agglomeration. The total pollutants from sources outside of the local region are still quantified but without specification of their location, using the method presented in Sect. 3.4.

- 5 If the goal is to provide source-receptor matrices for large regions (countries), then this method is probably not appropriate in its present form as the computational cost may be too high, and the level of detail provided is not needed. For such an application the method should be modified, so that the tracking is not done for individual grid cells, but for larger source areas or group of emission sources.

5.2 Downscaling

- 10 One obstacle to combine fine scale (urban) and regional modelling is the problem of "double counting". In the regional scale model, there is usually only one total concentration value, without distinction between its origins. Distinguishing between urban and background pollution can be difficult in practice (Thunis, 2018),

Ideally, the regional model should only compute the background/regional contributions and the fine scale model can then add the local contribution. In a city, scales down to street level may be required. Those very fine scale models will not compute

15 accurately the transport between distant streets within the city and the regional model must account for those. But if the same emissions source are included both in the regional and fine scale model they will be accounted for twice.

The Local Fractions can give the relative contributions from different sources directly. Thus, it is possible to either redistribute or replace only the appropriate local contributions using the more accurate fine scale model. ~~This avoids several of the problems associated with quantifying sources of different origins (Thunis, 2018).~~

20 An example for an operational downscaling tool is "uEMEP" (= urban EMEP), which combines the method described in this paper with the EMEP MSC-W air quality model (Simpson et al. , 2012), to provide daily air quality forecasts for all of Norway (<https://luftkvalitet.miljostatus.no/> , Denby et al. , 2020).

5.3 Improved modelling

Concentration of pollutants near the surface are required to assess health impacts or dry deposition. However, in many CTMs, 25 the lowest layer is several tens of meters thick, and the concentrations of pollutants will have a non-constant vertical profile within the layer. The shape of the profile will depend on the local conditions: if the pollutants are emitted locally at the surface the concentration will typically decrease with height, while the opposite is true for background pollutants. With the knowledge of the Local Fractions it is possible to improve the description of the vertical profile, and thus a more accurate estimation of, for instance, 3 meter concentrations (useful for health impact studies) or dry deposition rates.

30 As shown in Fig. 1 and 2 the Local Fractions vary strongly in space and time. If this information can be used to give better estimations of vertical profiles of pollutants it should have a significant effect on the results.

5.4 Future work

In this work, sources are always defined in an individual grid cell. The relative position of the source, $(\Delta x_s, \Delta y_s)$, could be replaced by a generic index that would point to more general groups of grid cells or regions. The formalism would be the same, except that emissions from any grid cell from the relevant region should be added together in the Local Fraction. This would 5 allow for instance to distinguish individually all grid cells in the immediate vicinity of the receptor grid cell, and successively larger regions as the distance increases. Another application could be to define countries as emitter regions.

In the future we plan to generalize the method to also include chemical processes ~~.-The Local Fractions could then give information about sensitivities to changes in emissions without necessarily summing up to one hundred percent—in some simplified form. The ambition is to still provide information for a very large number of sources, but to describe chemical processes in an approximate way. Compared to existing tagging methods, it will trade accuracy for computational efficiency.~~ 10

Code and data availability. The full EMEP MSC-W model code and main input data are publicly available through a GitHub repository under a GNU General Public License v3.0 (name emep-ctm). The routines related to the Local Fractions are part of the standard model. The exact version of the model used to produce the illustrative examples used in this paper (rv4.33) is archived on Zenodo (doi: 10.5281/zenodo.3265912).

15 *Competing interests.* The authors declare that they have no conflict of interest.

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