REPLIES TO REVIEWER #2

We thank reviewer 2 for the insightful comments, and for pointing to inconsistencies. We apologize for needing more time than anticipated to address all comments, but we believe that we have been able to address most issues, and that we have significantly strengthened the manuscript.

Before addressing the comments we would like to mention that we have modified the abbreviation of the O_3 health exposure metric (6 monthly daily maximum 1-h concentration) from M6M to 6mDMA1 (and accordingly M3M to 3mDMA1) as the latter seems to be commonly used in other works.

In the following we have placed the numbered reviewer comments in boxes. Our reply to the reviewer is in blue font, the changes to the manuscript in red font.

We also attach a revised version of manuscript and supplement with tracked changes compared to the first version.

REVIEWER 2 comments:

The manuscript by Van Dingenen et al. presents and evaluates TM5-FASST, a reduced form air quality assessment tool. The manuscript is long, but does a thorough job of both presenting the computational methods used for formulating the FASST tool and the types of impacts that it calculates (from health impacts to climate) as well as evaluating the tool against simulations from the full TM5 model as well as results in the literature. I have some additional questions in a few areas, described below, but in general was satisfied / impressed with the evaluation and performance. The writing could use a bit more editing for grammar and some of the figures need clarification on units, axis, etc. Addressing these will amount to moderate revisions and some additional evaluations.

1) However my only main concern about would be if this article should be moved to GMD instead of ACP, as the emphasis really is on the tool development and evaluation; there is not any content on application of the tool to new science or policy questions. It may not thus fit the scope of ACP.

REPLY:
We agree that this paper would have been also suited for GMD. However, due to high relevancy of this publication for the work of the TF HTAP, we decided that thematically the paper also fitted very well in this ACP special issue. Unfortunately, for this special issue, it was decided not to have a joint special issue between GMD and ACP (or other Copernicus journals), which would have been the perfect solution. We are confident that the interested reader will also find this publication in ACP.

2) 2.1-8: Why are reduced form or source-receptor models needed in the first place? I think there's a significant point to be made here about the complexity of air quality modeling vs the level of sophistication and computational intensity that can be acceptable to the decision-making community. But the article as presently writing misses this point, so the justification for the tool isn't readily apparent

REPLY:
Thank you for making this point, it is indeed important to introduce these issues to a general readership. This comment is closely related to the next one, so we address both in the introduction which has been expanded.

CHANGES TO MANUSCRIPT: *Changed first part of the introduction to: (P2 L14)*

1. Introduction

A host of policies influence the emissions to air. In principle any policy that influences the economy and use of resources will also impact emissions into the atmosphere. Specific air pollution policies aim to mitigate the negative environmental impacts of anthropogenic activities, some of which may be affected by other policies, like climate mitigation actions, transport modal shifts or agricultural policies. Further, air quality policies may impact outside their typical environmental target domains (human and ecosystem health, vegetation and building damage,...) for instance through the role played by short-lived pollutants in the Earth's radiation balance (Myhre et al., 2011; Shindell et al., 2009). Insight into the impacts of policies in a multi-disciplinary framework through a holistic approach could contribute to a more efficient and cost-effective implementation of control measures (e.g. Amann et al., 2011; Maione et al., 2016; Shindell et al., 2012).

Several global chemical transport models are available for the evaluation of air pollutants levels from emissions, sometimes in combination with off-line computed climate relevant metrics such as optical depth or instantaneous radiative forcing (e.g. Lamarque et al., 2013; Stevenson et al., 2013). These models provide detailed output, but are demanding in terms of computational and human resources for preparing input, running the model, and analyzing output. Further they often lack flexibility to evaluate ad-hoc a series of scenarios, or perform swift what-if analysis of policy options. Therefore there is a need for computationally-efficient methods and tools that provide an integrated environmental assessment of air quality and climate policies, which have a global dimension with sufficient regional detail, and evaluate different impact categories in an internally consistent way. Reduced-form source-receptor models are a useful concept in this context. They are typically constructed from pre-computed emission-concentration transfer matrices between pollutant source regions and receptor regions. These matrices emulate underlying meteorological and chemical atmospheric processes for a pre-defined set of meteorological and emission data, and have the advantage that concentration responses to emission changes are obtained by a simple matrix multiplication, avoiding expensive numerical computations. Reduced-form source-receptor models (SRM) are increasingly being used, not only to compute atmospheric concentrations (and related impacts) from changes in emissions but they have also proven to be very useful in cost optimization and cost-benefit analysis because of their low computational cost (Amann et al., 2011). Further, because of the detailed budget information embedded in the source-receptor matrices, they are applied for apportionment studies, as a complementary approach to other techniques such as adjoint models (Zhang et al., 2015) and chemical tagging (e.g. Grewe et al., 2012).

Although the computational efficiency of SRMs comes at a cost of accuracy, regional detail and flexibility in spatial arrangement of emissions, they have been successfully applied in regional studies (Foley et al., 2014; Li et al., 2014; Liu et al., 2017; Porter et al., 2017) and have demonstrated their key role in policy development (Amann et al., 2011).

3) Intro: Overall the introduction is rather brief. There are other reduced form models on regional scales that are used for different purposes (in the US and Asia, in particular). There are also theoretical advantages (quick) and disadvantages (approximations of linearity; enforced aggregation at pre-defined scales; outdated emissions inventories or old atmospheric conditions) of reduced form models. A lot more thought could be put into discussion and introducing these issues. This is ACP, not GMD, so more than just a model description is expected

REPLY: see previous comment

4) 3.18: This sentence is a bit too vague to be useful. The authors should mention what type of model updates have been made (emissions? aerosols? etc) and why they are deemed to not be relevant for this current work.

REPLY: We have added some more text to this paragraph to explain the major differences. The choice of emissions is not relevant in this context as emission datasets are external to the model framework, and in general chosen by the user depending on scientific issue.

CHANGES TO MANUSCRIPT: *Added to Section 2.1 (P4 L24)*

TM5 results used in the present study allow comparison with a range of other global model results in HTAP1, but ignore subsequent updates and improvements in TM5 as for instance described in Huijnen et al. (2010), which we consider not critical for this study. The most recent TM5 model does no longer consider zoom regions, but recoded the model into a Massive Parallel framework, enabling efficient execution on modern computers. While global horizontal resolution (1°x1°) is similar to the resolution of the most refined zoom region in TM5, vertical resolution was increased. Further, the model also uses vertical mass fluxes from the parent ECMWF meteorological model, not available at the time of development of TM5-cy2-ipcc, which could lead to somewhat different mixing characteristics. The gas phase chemical module has been updated to a modified version of CMB5.

.

5) This does raise the question of uncertainties introduced in this tool owing to use of a single year (was 2001 an average year, in terms of temp, precipitation, etc.?) to approximate a reasonable climatology, as well as this use of a year that is significantly older than most present applications, considering decadal-scale climate change.

REPLY: This is an issue raised by both reviewers. We agree with the reviewer that the year 2001 meteorology is somewhat outdated. The perturbation runs for constructing the SR library of FASST were performed with the TM5 model set-up defined in the first phase of HTAP1 (during the period 2008 – 2011) and because of the computational costs, an update with more recent meteorology was not possible (TM5 is not taking part in HTAP2 where meteorological year 2010 has been used). A systematic check of the representativeness of this particular year for each of the FASST regions is beyond the scope of this study, in the first place because FASST is considered to be a screening tool focussing on impacts of emission changes. However we have substantially extended the discussion on the use of a single meteorological year.

CHANGES TO MANUSCRIPT: *Added to Section 2.1 P5 L10*

Meteorological fields are obtained from the ECMWF operational forecast representative for the year 2001. The implications of using a single meteorological year will be discussed in section 4.2.

Discussion section 4.2 added (P31 L25)

4.2 Inter-annual meteorological variability

A justified critique on the methodology applied to construct the FASST SRs relates to the use of a single and fixed meteorological year 2001, implying possible unspecified biases in pollutant concentrations and source-receptor matrices compared to using a 'typical meteorological/climatological year'. We followed the choice of the meteorological year 2001 made for the HTAP1 exercise. As the North-Atlantic Oscillation (NAO) is an important mode of the inter-annual variability in pollutant concentrations and long range transport (Christoudias et al., 2012; Li et al., 2002; Pausata et al., 2013; Pope et al., 2018), the HTAP1 expectation was that this year was not an exceptional year for long-rang pollutant transport - e.g. for the North-Atlantic region, as indicated by a North Atlantic Oscillation (NAO) index close to zero for that year (https://www.ncdc.noaa.gov/teleconnections/nao/). The HTAP1 report (Dentener et al., 2010) also suggested that “Inter-annual differences in SR relationships for surface O_3 due to year-to-year meteorological variations are small when evaluated over continental-scale regions. However, these differences may be greater when considering smaller receptor regions or when variations in natural emissions are accounted for”. The role of spatial and temporal meteorological variability can thus be reduced by aggregating resulting pollutant levels and impacts as regional and annual averages or aggregates, the approach taken in TM5-FASST. The impact of the choice of this specific year on the TM5-FASST model uncertainty or possible biases in base concentrations and SR coefficients is not easily quantified. For what concerns the pollutant base concentrations, some insights in the possible relevance of meteorological variability can found in the literature. For example, Anderson et al., (2007) showed that in Europe, the meteorological component in regional inter-annual variability of pollutant concentrations ranges between 3% and 11% for airborne pollutants (O_3, $PM_{2.5}$), and up to 20% for wet deposition. On a global scale, Liu et al. (2007) demonstrated that the inter-annual variability in PM concentrations, related to inter-annual meteorological variability can even be up to a factor of 3 in the tropics (e.g. over Indonesia) and in the storm track regions. A sample analysis (documented in section S2.2 of the SI) of the RCP year 2000 emission scenario with TM5 at 6°x4° resolution of 5 consecutive meteorological years 2001 to 2005 indicates a year-to-year variability on regional $PM_{2.5}$ within 10% (relative standard deviation) and within 3% for annual mean O_3. We find a similar variability on the magnitudes of 20% emission perturbation responses within the source region for 6 selected regions (India, China, Europe, Germany, USA and Japan). The relative share of source regions to the pollutant levels within a given receptor region shows a lower inter-annual variability (typically between 2 and 6% for $PM_{2.5}$) than the absolute contributions.

6) 4.29: There is extensive research on the chemical oxidation of elemental carbon and the role this plays on the lifetime of this species in the atmosphere. Comment on why this is not included.

REPLY: The referee is right, also primary pollutants can undergo chemical conversion – however we feel this comment relates rather to 6.15 where we state that in TM5 (and FASST) the lifetime of BC and POM is not changing. The statement in 4.29 was intended to point out the difference between primary and secondary pollutants where in the latter case a completely new chemical compound is formed from precursors via chemical reactions, while for primary pollutants, dispersion and deposition are the primary process affecting their atmospheric concentration. Since the development of TM5, in literature two approaches have been developed towards parameterizing 'ageing' of elemental carbon. Ageing through condensation of hydrophobic species such as SO4 (and in the real world also other soluble components) is considered in e.g. the HAM aerosol physics model (Stier et al., 2005). The second approach considers oxidation of carbonaceous aerosol by O_3 following Tsigaridis and Kanakidou (2003). More recent work (e.g. Huang et al., 2012) analyses the joint impact of the two approaches, explicitly including the chemical-physical ageing processes. In general including the explicit processes tends to lengthen the atmospheric residence time of EC/BC compared to the earlier simple parameterisation in CTMs. The reason of not including these processes at the time of the release of TM5-JRC-Cy2-IPCC was that at that time none of the approaches was robustly anchored in improved performance at multiple observational sites, while at the same time the uncertainties in the wet removal parameterization were (and still are) also highly uncertain.

CHANGES TO MANUSCRIPT*: We feel this comment addresses original 6.15 rather than 4.29*

Original 6.15

BC and POM emissions are assumed not to interact with other pollutants and their atmospheric lifetime are assumed not to 15 be affected by mixing with other soluble species like sulfate, nitrate or ammonium salts

modified to (P8 L22):
BC and POM are assumed not to interact with other pollutants and their atmospheric lifetimes are prescribed and assumed neither to be affected by mixing with other soluble species like sulfate, nitrate or ammonium salts, nor to undergo oxidation by O_3. Recent work (e.g. Huang et al., 2012) indicates that a parametrized approach, as applied in TM5, tends to underestimate BC and POM atmospheric lifetimes, leading to a low concentration bias. When explicitly modelled, including the combined impact of both mechanisms, Huang et al., 2012 find that the global atmospheric residence times of BC and POM are lengthened by 9% and 3% respectively.

7) 5.1: I'm not sure what is a "parameter" in this context – please explain.

REPLY: Thank you for pointing out, this is a typo that has been corrected

CHANGES TO MANUSCRIPT: replaced 'parameters' by 'pollutants'

8) 5.1 - 14: It seems like some discussion of the fact that this functional relationship is only approximate is warranted. Instead, it is presented here as if the actual functional relationship is known, where in fact just a local linear approximation is used. This must have some limitations. For example, what is the basis for the statement later on this page that -20% perturbation is small enough to evaluate sensitivities and large enough for extrapolation? I recognize that -20% is a commonly used modeling experiment, but it is also commonly known that this approach has limitations for source attribution that are well documented in the literature (compared to tagging, 2nd order methods, or other).

REPLY: The referee makes a good point here, obviously the linear approach is approximate and has both advantages and limitations. We have already addressed most of this discussion in the introduction. We also modified the relevant phrase in the text.

CHANGES TO MANUSCRIPT:

Replaced:
In the current version v0 of TM5-FASST the function is a linear relation expressing the change in pollutant concentration in the receptor region upon a change in precursor emissions in the source region…

By (P6 L27):
In the current version v0 of TM5-FASST the emission-concentration relationship is locally approximated by a linear function expressing the change in pollutant concentration in the receptor region upon a change in precursor emissions in the source region…

9) The notation in equations (1) and (2) is not correct. In equation 1, there is an inconsistency between the description of the notation for the concentrations vs emissions species (i and j) and what is written in the equation. Assuming the equation is correct, the text should refer to change in concentration of component j (not i) owing to emitted precursor i (not j).

REPLY: Indeed, thanks for spotting.

CHANGES TO MANUSCRIPT:
Replaced:

For each receptor point y (i.e. each model vertical level 1°x1° grid cell), the change in concentration of component i in receptor y resulting from a -20% perturbation of emitted precursor j in source region x, …

By (P7 L29):
For each receptor point y (i.e. each model vertical level 1°x1° grid cell), the change in concentration of component j in receptor y resulting from a -20% perturbation of emitted precursor i in source region x, …

10) In equation (2), the notation on the summations is not complete nor correct. The first sum should be from x = 1 (below the sum) to n_x (written above the sum), and the second should be for i = 1 (below the sum) to n_i(j) (above the sum). It's also not clear why y would be bold in this equation. As explained in the text, the number of precursor pollutants (n_i) depends on the pollutant response in consideration, hence n_i is n_i(j). So the pollutant responses are dry aerosol concentrations? At what T,P,RH?

REPLY:
Indexing has been corrected and bold face removed.
The stored SR matrices for each component are indeed the dry mass, as obtained from the TM5 model lower layer (or as column density for radiative properties), using the meteorological data for year 2001.
For comparison with measurements and for health impact assessment FASST provides an estimate of $PM_{2.5}$ residual H2O at 35% RH and 25°C using mass growth factors for ammonium salts of 1.27 (Tang, 1996) and sea-salt of 1.15 (Ming and Russell, 2001). This allows for a calculation of $PM_{2.5}$ mass simulating the protocol for determination of gravimetric $PM_{2.5}$ mass in monitoring networks, and these are also the values on which epidemiological studies are based. Radiative forcing obviously takes into account atmospheric RH conditions.

CHANGES TO MANUSCRIPT:
Added below Eq. (1) (P8 L1):
In the present version TM5-FASST_v0, the SR coefficients for pollutant concentrations are stored as annual mean responses to annual emission changes. Individual $PM_{2.5}$ components SRs are stored as dry mass (μg m^{-3}). $PM_{2.5}$ residual water at 35% is optionally calculated a posteriori for sensitivity studies, assuming mass growth factors for ammonium salts of 1.27 (Tang, 1996) and for sea-salt of 1.15 (Ming and Russell, 2001). The presence of residual water in $PM_{2.5}$ is not irrelevant: epidemiological studies establishing $PM_{2.5}$ exposure-response functions are commonly based on monitoring data of gravimetrically determined $PM_{2.5}$, for which measurement protocols foresee filter conditioning at 30 – 50% RH. As many health impact modelling studies consider dry $PM_{2.5}$ mass or do not provide information on the inclusion of residual water we use dry $PM_{2.5}$ for health impact assessment in this study for consistency, unless mentioned differently.

Correcting indexing (P8 L11):
The total concentration of component (or metric) *j* in receptor region *y*, resulting from arbitrary emissions of *all* n_i precursors *i* at *all* n_x source regions *x*, is obtained as a perturbation on the base-simulation concentration, by summing up all the respective SR coefficients scaled with the actual emission perturbation:

$$C_j(y) = C_{j,base}(y) + \sum_{k=1}^{n_x} \sum_{i=1}^{n_i} A_{ij}[x_k, y] \cdot [E_i(x_k) - E_{i,base}(x_k)] \qquad (2)$$

11) 6.21: This equation needs to be corrected following suggestions for equation (2).

REPLY: Correct, done.

12) 6.23: It is oxymoronic to refer to secondary biogenic POM. This would just be secondary biogenic OM.

REPLY: Apologies for the confusion, but in this case POM actually stands for particulate organic matter.

CHANGES TO MANUSCRIPT: *changed P4 L16 to* "Biogenic secondary organic aerosol (BSOA)"

13) 6.24: Can the authors comment on how neglect of anthropogenic SOA might be biasing the results of this tool?

REPLY:
This is a difficult question, which may be worthy of an entire review. The main reason for ignoring anthropogenic SOA at the time of development of TM5- cy2-ipcc was that in the version of the CMB4 chemical scheme implemented in the model, Benzene and toluene chemistry was not included, as it was considered of local importance. In addition reliable global inventories were not available. Having said this, the importance of anthropogenic SOA will strongly depend on local emission strength and atmospheric chemistry conditions. For instance a recent conducted in China (Hu et al., 2017) suggest that in summer biogenic SOA is larger in summer (75 %) than in winter (25 %) 5 and over 35 μg/m^3 in 4 Chinese cities.
A global modelling study by (Farina et al., 2010) based on the volatility approach suggests that SOA formation from monoterpenes, sesquiterpenes, isoprene, and anthropogenic precursors is estimated as 17.2, 3.9, 6.5, and 1.6 Tg yr^{-1}, respectively. While in that study global levels of SOA were low (annual average 0.02 ug/m3)- in particular in Europe and China levels up to 1 ug/m3 were calculated, where levels of primary organic aerosol were reaching 20 ug/m3. Although this back-off the envelop assessment suggest that for larger regions the impact is less than 5-10 %, in urban regions with high anthropogenic VOC emissions the impact may be larger.

CHANGES TO MANUSCRIPT:
We added following phrase to the discussion section 4.1 (P31 L17):

The omission of secondary organic PM in TM5 is estimated to introduce a low bias in the base concentration of the order of 0.1 μg m^{-3} as global mean however with regional levels in Central Europe and China up to 1 μg m^{-3} in areas where levels of primary organic matter are reaching 20 μg m^{-3} (Farina et al., 2010) indicating a relatively low contribution of SOA to total PM$_{2.5}$

14) 6.25: Just because the impacts are annual in nature doesn't mean the emissions contributions to the impacts are seasonally consistent. Surely the impact of NO$_x$ on ammonium nitrate and O$_3$ is quite different in different seasons; it's not clear why one would have access to this information but not use it.

REPLY: It is certainly true that there are seasonal differences in emission-concentration sensitivities. However, when relevant, these seasonal trends are implicitly included in the exposure metrics and impacts. Several metrics are in fact based on detailed temporal ozone trends, e.g. considering only the daily maximal hourly value, or hourly values exceeding a 40 ppb threshold during the crop growing season. These responses – seasonal in nature – are stored to be scaled with annual emissions. Health impacts from PM$_{2.5}$ are based on annual averaged values and are not evaluated on a seasonal basis. Hence, although there may be scientific (process understanding) interest in elaborating seasonal trends, from a health/crop/climate impact assessment perspective, there is not much added value storing temporal trends in the source-receptor matrices which would come at a high computational cost (multiplying the number of SR matrices with 12).

15) 7.7-21: I got a bit lost in this discussion of the way CH4 concentrations responses are treated. It would be good if this section could be expanded and formalized a bit better, using equations where useful, such that the approach could be evaluated and replicated.

REPLY: This was indeed not explained in an optimal way. As there are two instances in the paper where CH4 responses are treated (O_3 response from CH4 emissions, and indirect forcing from short-lived precursors on CH4 and background ozone – see next comment) we have moved and expanded the description of the methodology, which is based on our interpretation of published work, in the SI (S3)

CHANGES TO MANUSCRIPT: added *section S3 in the SI:*

S3.1 CH_4 – O_3 source-receptor relations from HTAP1 perturbation experiments:

CH_4 emissions lead to a change in CH4 concentrations with a perturbation response time of about 12 years. In order to avoid expensive transient computations, HTAP1 simulations SR1 and SR2 with prescribed fixed CH4 concentrations (1760 ppb and 1408 ppb, see Dentener et al., 2010) were used to establish CH4 – O_3 response sensitivities. Previous transient modeling studies have shown that a change in steady-state CH_4 abundance can be traced back to a sustained change in emissions, but the relation is not linear because an increase in CH_4 emissions removes an additional fraction of atmospheric OH (the major sink for CH_4) and prolongs the lifetime of CH_4 (Fiore et al., 2002, 2008; Prather et al., 2001).
In a steady-state situation, the CH4 concentration is the result of balanced sources and sinks. In the HTAP1 experiments, keeping all other emissions constant, the change in the amount of CH4 loss (mainly by OH oxidation with a lifetime of ca. 9 years, neglecting loss to soils and stratosphere with lifetimes of ca.160 and 120 years respectively (Prather et al., 2001)) under the prescribed change in CH4 abundance should therefore be balanced by an equal and opposite source which we consider as an “effective emission”. The amount of CH4 oxidized by OH in one year being diagnosed by the model, the resulting difference between the reference and perturbation experiment of -77 Tg sets the balancing “effective” emission rate to 77Tg/yr, which is then used to normalize the resulting O_3 and O_3 metrics response to a CH4 emission change.
The same perturbation experiments also allow us to establish the CH4 self-feedback factor F describing the relation between a change in emission and the change in resulting steady-state concentration:

$$\frac{C_2}{C_1} = \left(\frac{E_2}{E_1}\right)^F \qquad \text{(S3.1)}$$

With CH4 concentrations prescribed, CH4 emissions were not included in the SR1 and SR2 experiments. The feedback factor F is derived from model-diagnosed respective CH4 burdens (B) and total lifetimes (LT) as follows (Fiore et al., 2009; Wild and Prather, 2000):

$$F=1/(1-s) \qquad \text{(S3.2)}$$

$$s = \partial \ln(LT) / \partial \ln(B) \qquad \text{(S3.3)}$$

TM5 returns s = 0.33 which can be compared to a range of values between 0.25-and 0.31 in IPCC-TAR (Prather et al., 2001, Table 4.2) , resulting in a TM5-inherent calculated feedback factor F=1.5. This factor can be used to estimate the corresponding SR2-SR1 change in CH4 emission in a second way. From Eq. S3.1 we find that a 20% decrease in CH4 abundance corresponds to a 14% decrease in total CH4 emissions. Kirschke et al. (2013) estimate total CH4 emissions in the 2000s in the range 550 – 680 Tg yr-1 from which we obtain an estimated emission change between the HTAP SR1 and SR2 experiments in the range 77 – 95 Tg yr-1, in line with our steady-state loss-balancing approach.

16) It also wasn't clear to me – is NO_x allowed to impact CH4, particularly for the purposes of climate impacts?

REPLY: It is, as are all short-lived ozone precursors. We have added an extensive description in the SI on how the emission – forcing contributions in terms of (1) direct O_3 (2) indirect CH4 and (3) CH4-induced long-term O_3

CHANGES TO MANUSCRIPT: *added section S6.2 to the SI*

S6.2 Secondary forcing feedbacks of O_3 precursors on CH_4 and background O_3

Emissions of short-lived species (NO_x, NMVOC, CO, SO_2) influence the atmospheric OH burden and therefore the CH_4 atmospheric lifetime, which in turn contributes to long-term change in CH4 and background ozone. Hence, the total forcing contribution from O_3 precursors consists of a short-term direct contribution from immediate O_3 formation (S-O_3), and secondary contributions from CH_4 (I-CH4) and a long-term feedback from this CH_4 on background O_3 (M-O_3). We apply the formulation by (Fiore et al., 2009; Prather et al., 2001; West et al., 2007) to calculate the secondary change in steady-state CH_4 from SLS emissions, using the TM5 perturbation experiments for FASST (see section S3). TM5 diagnoses the CH_4 loss by oxidation for reference and perturbation run (where the emissions of SLS are decreased with -20%), from which we calculate the CH_4 oxidation lifetime ratio between reference and perturbation:

$$\frac{LT_P}{LT_{Ref}} = \frac{CH4\_ox_P}{CH4\_ox_{Ref}} \qquad [S6.5]$$

Where LT is the CH4 lifetime against loss by OH oxidation, and $CH4\_ox$ = the amount (Tg) of CH4 oxidized.
The new steady-state methane concentration M due to the changing lifetime from perturbation experiment P, induced by O_3 precursor emissions follows from (Fiore et al., 2008, 2009; Wild and Prather, 2000):
$M = M_0 \times \left(\frac{LT_P}{LT_{ref}}\right)^F$ where $M_0 = 1760$ ppb, the reference CH4 concentration and F = 1.5, determined from the HTAP1 CH4 perturbation experiments, as described in section S3.

The change in CH4 forcing (I-CH4) associated with the change to the new steady-state concentration is obtained from IPCC AR5 equations:

$$\Delta F = \alpha\left(\sqrt{M} - \sqrt{M_0}\right) - \left(f(M, N_0) - f(M_0, N_0)\right) \qquad [S6.6]$$
$$f(M, N) = 0.47 ln[1 + 2.01 \times 10^{-5}(MN)^{0.75} + 5.31 \times 10^{-15} M(MN)^{1.52}] \qquad [S6.7]$$
Where M, M0 = CH4 concentration in ppb, N0 = N2O (=320 ppb)

The associated long-term O_3 forcing (M-O_3) per Tg precursor emitted is obtained by scaling linearly the change in O_3 forcing obtained in the HTAP1 CH4 perturbation simulation (SR2–SR1), with the change in CH4 obtained above, and normalizing by the precursor emission change (Fiore et al., 2009)

$$\Delta F = \frac{\Delta F_{O3[SR2-SR1]}}{M_{SR2} - M_{SR1}} (M - M_0) \qquad [S6.8]$$

The response of CH4 and O_3 forcing to CO emission changes (for which no regional TM5-FASST perturbation model simulations were performed) was taken from TM5-CTM simulations performed for the HTAP1 assessment (Dentener et al., 2010) using the average forcing

efficiency for North America, Europe, South-Asia and East-Asia. For regions not covered by the HTAP1 regions, the HTAP1 rest-of-the-world forcing efficiency was used.
The resulting region-to-globe emission-based forcing efficiencies are given in Tables S6.2 to S6.5 for aerosols, CO, CH4 and other O_3 precursors respectively.

17) Many of the studies in the table are a bit out-of-date, as they would be around atmospheric conditions / emissions levels that are rather old, or in comparison to datasets that have greatly matured (for example comparison to satellite-based NO2 retrievals, which are now much more accurate and consistent across retrievals than in the study of van Noije 2006.

REPLY: This is indeed the case. However, the TM5 version used in this study was developed and evaluated in the studies shown in the table. Since then no new developments and evaluation studies have been performed on the version used in this work. As in this study we are focusing on an evaluation of TM5-FASST, using TM5 as a reference, it is beyond the scope of this study to re-evaluate TM5 with new data sets, which would be worth one or more new papers on its own.

CHANGES TO MANUSCRIPT: *added following phrase to section S2.1 in the SI:*

We are aware of recent more accurate observational data have become available for the validation of the model since the validation studies listed in Table S2.1, in particular from satellite-based retrievals. However here we focus on the validation of FASST, using TM5 as a reference, and it is beyond the scope of this study to re-evaluate the TM5 model itself.

18) 8.3-4: Statements like this could be supported by reference many articles on the topic, including evaluation of how much this matters for different species at different scales.

REPLY:
We agree with this point. We address this now in a dedicated section in the discussion where we refer to exemplary studies that have specifically addressed the issue of grid resolution on exposure. They indicate in general that O_3 tends to be overestimated and (primary) $PM_{2.5}$ tends to be underestimated compared to higher resolution models. Further we have included in the SI a quick analysis of the TM5 base simulation at resolution 6°x4°, 3°x2° and 1°x1° to illustrate the impact of resolution on concentrations and emission-concentration response sensitivities.

CHANGES TO MANUSCRIPT:

Modified last part of section 2.1 (P5 L28):

The model grid resolution influences the predicted pollutant concentrations as well as the estimated population exposure, especially near urban areas where strong gradients occur in population density and pollutant levels, which cannot be resolved by the 1°x1° resolution. In section 2.4 we describe a methodology to improve population $PM_{2.5}$ exposure estimates by applying sub-grid concentration adjustments based on high-resolution ancillary data. The bias introduced by model resolution affects as well computed SR matrices, e.g. off-setting the share of 'local' versus 'imported' pollution in a given receptor region. We will discuss this aspect more in detail in section 4.3.

Added section 4.3 to the discussion section (P32 L20):

4.3 Impact of the native TM5 grid resolution on pollutant concentration and SRs

FASST base concentrations and SRs have been derived at a 1°x1° resolution which is a relatively fine grid for a global model, but still not optimal for population exposure estimates and health

impact assessments. Previous studies have documented the impact of grid resolution on pollutant concentrations. The effect of higher grid resolution in global models is in general to decrease ozone exposure in polluted regions and to reduce O_3 long-range transport, while $PM_{2.5}$ exposure – mainly to primary species - increases (Fenech et al., 2018; Li et al., 2016; Punger and West, 2013). Without attempting a detailed analysis, a comparison of TM5 available output for $PM_{2.5}$ and O_3 at 6°x4°, 3°x2° and 1°x1° resolution confirms these findings, as illustrated in Fig. S2.6 of the SI. Although FASST is expected to better represent population exposure to pollutants than coarser resolution models, a resolution of 1°x1° may not adequately capture urban scale pollutant levels and gradients when the urban area occupies only a fraction of the grid cell. The developed sub-grid parameterization for $PM_{2.5}$, providing an order-of-magnitude correction which is consistent with a high-resolution satellite product, is subject to improvement and to extension to other primary pollutants (NO_2, e.g. Kiesewetter et al., 2014, 2015) and O_3. To our knowledge a workable parametrization to quantify the impact of sub-grid O_3 processes on population exposure – in particular titration due to local high NO_x concentrations in urban areas - has not been addressed in global air quality models.
The impact of grid resolution on the within-region source-receptor coefficients can be significant, in particular for polluted regions where the coarse resolution includes ocean surface, like Japan. Table S2.3 in the SI shows as an example within-region and long-range SR coefficients for receptor regions Germany, USA and Japan. A higher grid resolution increases the within-region response and decreases the contribution of long–range transport (where the contribution of China to nearby Japan behaves as a within-region perturbation). In the case of Japan, the within-region $PM_{2.5}$ response magnitude increases with a factor of 3, and the sign of the within-region O_3 response is reversed when passing from 6°x4° to higher resolution. Also over the USA, the population-weighted within-region response sensitivity upon NO_x perturbation increases with a factor of 5. Further, we find that in titration regimes, the magnitude of the O_3 response to NO_x emissions increases with resolution (i.e. ozone increases more when NO_x is reduced using a fine resolution) while the in-region ozone response is reduced in non-titration regimes (India and China, Fig. 2.7d). These indicative results are in line with more detailed studies (e.g. Wild and Prather, 2006).

19) Is there a reason why primary $PM_{2.5}$ from industrial sources would also not be expected to contribute to the local urban increment? Or is this source just not very large?

REPLY: The choice of including only transport and residential sectors contributing to the urban increment is mainly motivated by the fact that these are the sectors for which the emissions correlate best with urban population. In developed countries, industrial emissions are typically somewhat away from densely populated areas in city centres, and elevated stack heights avoid direct exposure of the population. Nevertheless we recognize that depending on the local context, and especially in developing countries, these conditions may not or not completely be fulfilled. We further note that the intercomparison with satellite data (S4 in the SI) seems to indicate that the present sub-grid incremental factor is already at the high side, and including more industrial sources for primary $PM_{2.5}$ increments would worsen the bias.

CHANGES TO MANUSCRIPT:

Modified the original phrase
Indeed, secondary $PM_{2.5}$ is formed over longer time scales and therefore deemed to be more homogeneously distributed at the regional scale

To (P10 L23):
Indeed, secondary $PM_{2.5}$ is formed over longer time scales and therefore deemed to be more homogeneously distributed at the regional scale, while primary $PM_{2.5}$ emissions from other

sources than the residential and transport sector are assumed to occur more remotely from urban areas.

20) Section 2.4 and SIS4 are useful in understanding the urban increment, and some evaluation of improvement in performance compared to satellite-derived $PM_{2.5}$ is included. However, the evidence is a bit indirect. I'd like to see a comparison of native 1x1 and urban downscaled BC concentration to in situ measurements from urban monitoring sites, such as are available in the US.

REPLY: The referee is correct in stating that the evidence is indirect and that improvements are possible. However we feel that an intercomparison with BC from monitoring stations is not the most appropriate way, because

- there are large uncertainties with BC mass measurements
- BC in TM5 really represent Elemental Carbon (excluding observation that are based on optical measurements),
- not in the least TM5, like many other models, has a low-bias towards observations.

Further, the urban-incremented FASST mean 1°x1° concentration is not directly comparable to point measurements of monitoring stations in particular when placed in urban locations. To address the reviewer's comment, we have instead elaborated the recent data set of van Donkelaar et al. (2016) which integrates a $PM_{2.5}$ satellite product for anthropogenic $PM_{2.5}$ with data from monitoring stations. The data set is available at a 0.1°x0.1° resolution, allowing for an aggregation at population-weighted 1°x1° grid mean that can directly be compared to FASST native as well as urban-incremented concentrations at grid cell or regional level.

CHANGES TO MANUSCRIPT:
We have significantly extended section S4 of the SI with additional text, figures and tables. We include here the new text of the section and refer to the revised SI for the figures.

S4.2 Comparison of TM5-FASST urban incremented $PM_{2.5}$ with observations

We use the year 2010 0.1°x0.1° resolution global satellite product from the Dalhousie University Atmospheric Composition Analysis group (available at http://fizz.phys.dal.ca/~atmos/martin/?page_id=140), which includes ground-based observations via a Geographically Weighted Regression, while mineral dust and seasalt have been removed, as described in van Donkelaar et al., (2016).
The high-resolution satellite data (SAT) contain the sub-grid population and concentration gradients that we try to simulate with parametrization described above. Creating a SAT population-weighted average at 1°x1° resolution makes it possible to evaluate the TM5-FASST native and urban-incremented 1°x1° output. We convert the 0.1°x0.1° SAT resolution to the 2.5'x2.5' resolution of the CIESIN (year 2000) population dataset i.e. 24 sub-grid cells for each 1°x1° cell, to be overlaid with the satellite dataset. FASST $PM_{2.5}$ 1°x1° grid maps are calculated from the HTAP2 year 2010 emission inventory, includig the GFED v3 biomass burning emission inventor (REF). To remain consistent with the SAT product, residual water at 35% has been included. Fig. S4.1 shows global gridmaps of FASST and SAT $PM_{2.5}$ with dust and sea salt removed), and with the sub-grid increment included in the FASST result.
We evaluate both FASST native and urban incremented 1°x1°grid cell concentrations, using the parameterization described in the previous section. We calculate the following 1°x1° grid mean concentrations from the 2.5'x2.5' SAT $PM_{2.5}$ and population sub-grid cells

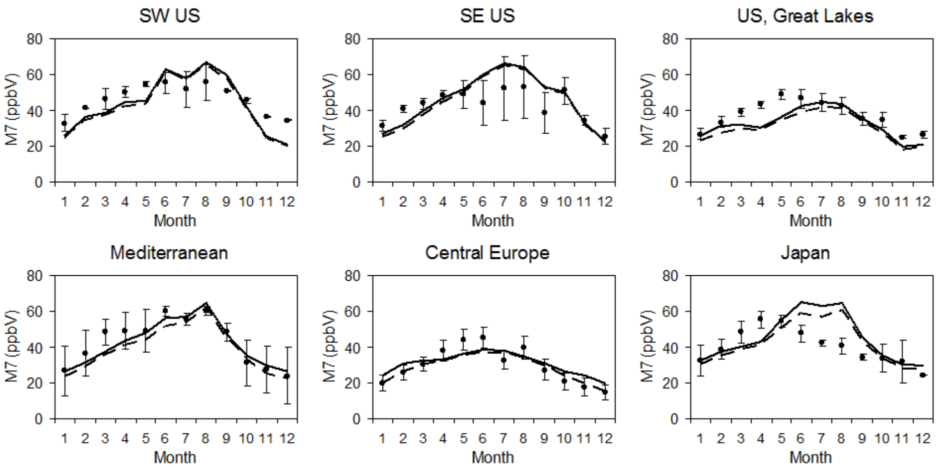
$$SAT_{AREA} = \frac{1}{24}\sum_{i=1}^{24} PM_{2.5,i}$$

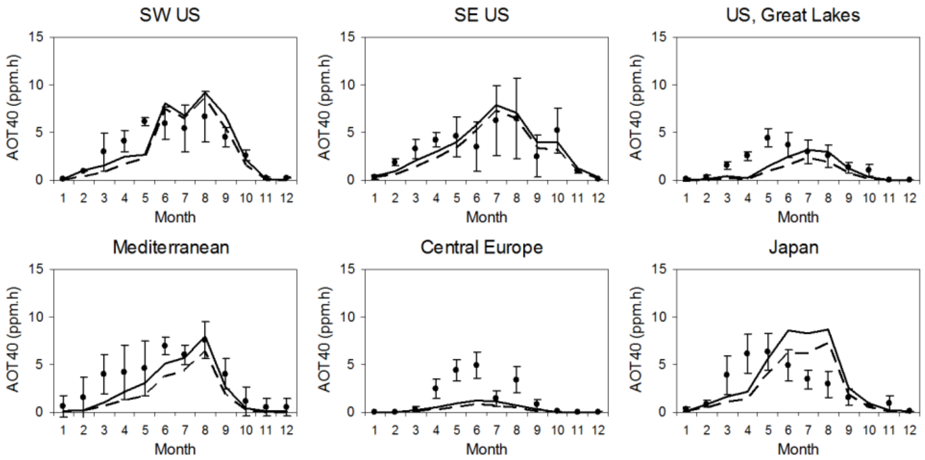
$$SAT_{POP} = \frac{\sum_{i=1}^{24} PM_{2.5,i} \cdot POP_i}{\sum_{i=1}^{24} POP_i}$$

SAT_{AREA} is the equivalent of the native FASST 1°x1° grid cell concentration, while SAT_{POP} represents the population-weighted mean 1°x1° concentration considering sub-grid gradients, to be compared with the FASST urban-incremented value, hereafter referred to as incremented concentrations. Regional and global mean population exposure to $PM_{2.5}$ (Table S4.3) is calculated using population-weighing on the 1°x1° grid cells, for both native (area-mean) and incremented concentrations.
Table S4.3and Fig. S4.2 show that for all regions, except for MEA (Mediterranean + Middle East), we find an over-all good agreement in regional mean $PM_{2.5}$ exposure between FASST and SAT, both for the native and incremented values. Figure S4.3 shows the absolute regional-mean increment in $PM_{2.5}$ exposure. We find that applying the FASST sub-grid parameterization increases global mean exposure with 1.4 µg m^{-3} (FASST), versus an increase of 1.1 from SAT, corresponding to a global population-weighted mean 5% increase for both methods. The FASST urban increment parameterization generates a regional-mean increase in $PM_{2.5}$ exposure from 0.6 µg/m^3 (Latin America) to 3.4 µg/m^3 (Russia and former Soviet Union states). In Europe and North-America the regional increase is around 1µg/m^3. Except for East-Asia and Latin America, the regional FASST increment exceeds the SAT value. SAT regional increments range between 0.3 µg/m^3 for Russia and former Soviet Union states and 1.8 µg/m^3 in East-Asia. Although we don't find a direct correlation between the SAT and FASST computed increments, it is encouraging that without applying any fitting procedure, and using two completely different approaches, increments from FASST and SAT are in the same order of magnitude.
Figs. S4.4 (Europe and North-America), S4.5 (China and India) and S4.6 (Africa and Latin America) show a detailed grid-to-grid comparison for selected key regions between native and incremented FASST on the one hand and SAT_{POP} on the other. In general, individual grid cells are reproduced within a factor of two. The FASST increment parameterization slightly improves the correspondence with SAT_{POP} compared to the native data except for China where the native FASST concentrations already exceed SAT_{POP}. Although an agreement at grid cell level is not the ambition of FASST, these results indicate that our crude approach is roughly performing, but that a more sophisticated approach in the urban increment may be warranted.
Finally, seen the large uncertainties on absolute $PM_{2.5}$ concentrations, one may wonder if the implementation of an urban increment parameterization is worth the effort. A FASST RCP2000 analysis of global mortalities with and without the generic urban increment factors (given in Table S4.2) shows that the global 5% increase in $PM_{2.5}$ exposure due to the urban increment accounts for an increase in total mortality numbers with 14% when dry $PM_{2.5}$ is considered, and with 11% when $PM_{2.5}$ is humidified at 35% RH. The difference is due to the threshold in the exposure-response functions (see section S5 in this SI). In areas where the native grid concentration is just below threshold, a small increase in $PM_{2.5}$ will have a strong response in mortalities while areas with native 1°x1° concentrations above the threshold will respond more proportional to the subgrid increment. Including hygroscopic growth at 35% from the onset reduces the cases where native resolution $PM_{2.5}$ remains below the threshold which explains the lower impact of the subgrid increment factor.

21) 11.25-27: This justification would be improved if the authors were a bit more quantitative. Also, if the lowest model level O_3 compares favorably to the surface O_3 measurements, this begs the question then of why the modeled O_3 thought the lower atmosphere in TM5 is biased low (as surface-level concentrations would be lower than 30 m concentrations).

REPLY: As can be seen in the paper by Van Dingenen et al. (2009), which uses the same TM5 model versions, but slightly different emissions (see their figures 6 and 7 copied below, but not used in the current manuscript) during the summer months (i.e. crop growing season), the daytime a vertical gradient beween 30 m (model centre) and 10 m (standard height of observations) is nearly absent – presumably due to higher atmospheric instability. The TM5 value at 30m is generally reproducing well the observations, and when it does not, the vertical gradient in TM5 is not the dominant factor causing discrepancies.





CHANGES TO MANUSCRIPT:

Changed phrase

However comparing TM5 simulated gridbox-centre ozone metrics with observations from 99 monitoring stations 25 worldwide, Van Dingenen et al. (2009) find that, averaged over the horizontal resolution of the grid cells, the TM5 simulated 30m monthly O_3 and O_3 metrics represent the observed values within their variability range.

To (P13 L10):

However comparing TM5 simulated gridbox-centre ozone metrics with observations from 99 monitoring stations worldwide, Van Dingenen et al. (2009) find that, when averaged at the regional scale, TM5 simulated crop metrics obtained from the grid box centre are reproducing the observations within their standard deviations, and that the monthly 10m TM5 metric values do not significantly improve the bias between model value and observations. Therefore we use the standard model output at 30m.

22) 12.21: The authors evaluate their approach to calculating direct aerosol radiative forcing by providing plots of the species specific forcings in Fig S6.1 and noting they are “reliable results.” However, I don’t have any sense of what makes these results reliable. What features of the distributions shown in these plots are those that we would expect, easily explain, or could compare to observations or other modeling studies? The BC RF in the eastern part of Antarctica exhibits a strange horizontal strip that I’m not sure about. Also, the figure legend on S6.1 is redundant and the units on both of the color bars are incorrect.

REPLY:
The reviewer correctly questions our use of the word reliable, which overstates our confidence in the uncertainty associated with the whole computational chain from emissions to concentrations and aerosol columns, and the scaling with normalized radiative forcing patterns. All these steps come with intrinsic uncertainties, and the radiative forcing uncertainty at grid basis is inevitably associated with relatively large uncertainties. The statement on 'reliability' is based on comparison with other globally aggregated results with an independent study performed by Unger et al, which gives remarkably similar source specific RF results. The 'strip' at the South Pole is likely due a numerical issue related the polar singularity and the necessary grid-size inflation in TM5 to deal with the singularity in a lon-lat projection.

CHANGES TO MANUSCRIPT: *Modified the phrase to (P14 L19):*

Our evaluation of pre-industrial to present radiative forcing in the validation section demonstrates that, in the context of the reduced-form FASST approach, the applied method however provides useful results. Figure S6.1 (a, b, c) in the SI shows the resulting global radiative forcing fields for sulfate, POM and BC. The regional emission-normalized forcing SRs for aerosol precursors (in W m^{-2} Tg^{-1}) are given in Table S6.2 of the SI.

Figure S6.1 has been modified to display the proper legend next to each graph, and units have been corrected to W mg^{-1}.

23) Section 2.7.2: The tool does not include the substantial non-direct cloud interactions for BC, nor the impact of BC on snow/ ice albedo. These factors contribute significantly to the targeting of BC-rich sources for SLCP mitigation. Comment on how omission would affect TM5-FASST results.

REPLY:
This is a correct observation, and indeed worth mentioning. Surface albedo effects (snow and sea-ice) is estimated to contribute with (+0.04 to +0.33) W/m^2, cloud interaction with (-0.47 to +1.0) W/m^2 on a total estimated forcing of (0.17 to 2.1) W/m^2 (Bond et al., 2013) where FASST estimates a total anthropogenic BC forcing of +0.15 W/m^2 hence all these contributions are significant. As mentioned in the conclusions, future developments could indeed include these effects, in particular changes in the surface albedo, seen the fact that BC deposition is computed by FASST. Nevertheless, we not that TM5, like other global models, has large uncertainties associated with the calculation of BC depositions.

CHANGES TO MANUSCRIPT:
In section 2.7.1 (P14 L15):
Neglecting the aerosol mixing state and using column-integrated mass rather than vertical profiles introduces additional uncertainties in the resulting forcing efficiencies. Accounting for internal mixing may increase the BC absorption by 50 to 200% (Bond et al., 2013), while including the vertical profile would weaken BC forcing and increase SO_4 forcing (Stjern et al. 2016). Further, the BC forcing contribution through the impact on snow and ice is not included, nor are semi- and indirect effects of BC on clouds. Our evaluation of pre-industrial to present

radiative forcing in the validation section demonstrates that, in the context of the reduced-form FASST approach, the applied method however provides useful results.

In section 3.3.1(P24 L25):
However, comparing to another widely used literature source (Bond et al., 2013), the TM5-FASST_v0 BC forcing estimate still falls within the 90% CI (0.08, 1.27) W/m^2 direct radiative forcing given for the year 2005, with a comparable global BC emission rate. Our low-end BC forcing estimate can be partly explained by the simplified treatment as externally mixed aerosol, without accounting for the enhancement of the mass absorption cross-section when BC particles become mixed or coated with scattering components. Not-included snow albedo and indirect cloud effects would contribute with +0.13 (+0.04 to +0.33) W/m^2 and +0.23 (-0.47 to +1.0) W/m^2 respectively (Bond et al., 2013).

24) Fig 2: What is the mechanism by which the perturbation in NO_x emissions causes a reduction in SO4 in IND (as opposed to an increase in all other regions)?

REPLY: This is indeed an interesting observation, which could be linked to the oxidative capacity of the atmosphere in that region, and/or to the thermodynamic properties of the ammonium-sulfate-nitrate system and the specific meteorological conditions in that area. Indeed, India has the particular feature that sulfate is dominating the inorganic aerosol fraction, and NH3 may be in excess. Answering this question would require a deeper analysis of TM5 budget data and the particular thermodynamic aerosol regimes for this case, where we notice that especially above India there are no reliable observations that could shed light on model discrepancies . Therefore we think that further analysis of this interesting model result is beyond the scope of this work where we focus on documenting and validating the linearity approach of FASST. However, in the text we point to this results for further multi-model analysis.

CHANGES TO MANUSCRIPT: *the paragraph has been expanded as follows: (P19 L4)*

For India we further observe a relative strong nitrate response to NO_x emissions, with NO_3^- increasing by a factor of 3 for a doubling of NO_x emissions, although the responses shown in Fig. 2 indicate that absolute changes (in μg m^{-3}) in NO_3 are relatively low and that secondary $PM_{2.5}$ in this region is dominated by SO_4. We are not aware of reliable observations or other published NO_x-aerosol sensitivity studies from that region that could corroborate this calculated sensitivity. Because such a feature may strongly affect projected future $PM_{2.5}$ levels and associated impacts, we recommend regional multi-model studies devote attention this feature.

25) 18.1-2: It seems NO_x levels in the US and Europe are much lower now, and I'm not sure these titrations still exist; they are at least less persistent in the summer. See for example recent article by Jin, Fiore, et al., JGR, 2017.

REPLY: Thank you for pointing to this interesting paper. Indeed NO_x emissions have been decreasing in the last two decades and indeed, the FASST SR relations were established for year 2000 conditions favouring a NO_x-saturation regime over W-Europe and NE-US. The fixed O_3 emission-response slopes are a major caveat for the evaluation of future scenarios, however, as already pointed out in the paper, while annual O_3 displays the typical reverse NO_x-O_3 response because of the winter-time titration, the slope reverses to positive in most cases when considering seasonal metrics centred on summer (Figure 6 in our paper). This being said, further reduction in NO_x and NMVOC is likely to change the O_3 (metric) response sensitivity, and indeed the fixed and linear SRs are a limitation of the tool. A possible, but non-trivial implementation, way to address this trend is to introduce higher order terms in the SRs and/or to update the year 2000 SRs with more recent ones obtained in the frame of HTAP2 (e.g. based on Turnock et al., 2018).

CHANGES TO MANUSCRIPT:
This issue is now introduced in the discussion section 4.1 (P30 L4)

The reliability of the model output in terms of impacts depends critically on the validity of the linearity assumption for the relevant exposure metrics (in particular secondary components), which becomes an issue when evaluating emission scenarios that deviate strongly from the base and -20% perturbation on which the current FASST SRs are based. The evaluation exercise indicated that non-linearity effects in $PM_{2.5}$ and O_3 metrics in general lead to a higher bias for stringent emission reductions (towards -80% and beyond) than for strong emission increases compared to the RCP2000 base case, but over-all remain within acceptable limits when considering impacts. Indeed, because of the thresholds included in exposure-response functions, the higher uncertainty on low (below-threshold) pollutant levels from strong emission reductions has a low weight in the quantification of most impacts. In future developments the available extended-range (-80%, +100%) emission perturbation simulations could form the basis of a more sophisticated parameterization including a bias correction based on second order terms following the approach by Wild et al. (2012) both for O_3 and secondary $PM_{2.5.}$ The break-down of the linearity at low emission strengths is relevant for O_3 and O_3 exposure metrics as the implementation of control measures in Europe and the US has already substantially lowered NO_x levels over the past decade, gradually modifying the prevailing O_3 formation regime from NO_x-saturated (titration regime) to NO_x-limited (Jin et al., 2017).

26) 18.6: The statement that the sensitivities of impact-relevant O_3 metrics (M6M and M12) are more linear than for annual average pop-weighted O_3 is not supported by the results shown in Fig 6. The responses to NO_x emission changes seem to be more nonlinear for M6M or M12 in some cases, such as GBR as well as others. The text should be revised accordingly.

REPLY: This is indeed wrongly formulated; this should rather refer to the changing sign of the slope.

CHANGES TO MANUSCRIPT: *changed phrase*
However, the impact-relevant O_3 metrics, both health and crop related, are based on summertime and daytime values and are expected to behave more linearly (Wu et al., 2009).

To (P20 L27):
On the other hand, the impact-relevant O_3 metrics, both health and crop related, are based on summertime and daytime values and are expected to be less affected by titration and consequently to maintain a positive emission-response slope (Wu et al., 2009).

27) 18.20: AOT40 would focus on high O_3 values. It's not clear to my, chemically speaking, why this would be expected to response more nonlinearly than other metrics. Presumably larger O_3 values are occurring more in the summer. Earlier it was claimed that summer sensitivities would be more linear … so I'm a bit confused here.

REPLY:
AOT40 is a threshold-based metric accumulating only values above 40ppb, and this built-in step function makes it difficult to approximate it with a linear function over a large perturbation range. For instance, in regions where ozone levels are just above 40ppb, a small decrease in O_3 can cause a big decrease in AOT40, while a similar small increase would cause a smaller AOT40 response (in absolute terms). Similarly, a SR sensitivity established from a perturbation at high O_3 levels will behave rather linearly in the high O_3 range, but cannot be extrapolated to very

strong reductions where it will lead to an overestimation of AOT40. Therefore the SR sensitivity based on the 20% decrease is less likely to be generally applicable over a large perturbation range.
This is less the case of metrics like M12 which are based on 3-monthly means of daytime ozone and behave more linearly with respect to emission perturbations.

28) 18.31: Why is that the case, chemically speaking?

REPLY: In this case – without investigating the underlying chemical mechanism - the larger deviation is a consequence of the slight convex shape of the O_3 response to NO_x for these countries, combined with the extrapolation of the -20% slope to larger perturbations.

CHANGES TO MANUSCRIPT:
With the major revision of sections 3.1.2 this particular phrase and the figures it was referring to have been removed. However we do mention (P19 L10):

Because the TM5-FASST linearization is based on the extrapolation of the -20% perturbation slope, concave-shaped trends in Fig. 3 indicate a tendency of TM5-FASST to over-predict secondary $PM_{2.5}$ at large negative or positive emission perturbations, and opposite for convex-shaped trends.

29) 20.4-14: I find it interesting that the change in $PM_{2.5}$ is predicted by FASST better than absolute concentrations (which I would expect) but that the change in O_3 metrics is predicted more poorly than absolute concentrations (would not expect). Do the authors have any thoughts about the reasons behind the latter?

REPLY:
Ozone behaves in general less linearly than $PM_{2.5}$, i.e. the perturbation term in Eq. 2 is more robust for $PM_{2.5}$ than for O_3. For strong perturbations, either side of the reference case, total $PM_{2.5}$ is overpredicted, hence making the difference between the high and low emission case cancel out some of the bias compared to absolute total $PM_{2.5}$.
For O_3, one must consider that the relative contribution of the “base” term in Eq. 2 is relatively high, even for strong anthropogenic perturbations because it contains the natural background. Roughly speaking, setting all anthropogenic emissions to 0 would still leave about 30 ppb of 6mDMA1. Therefore, this fixed contribution in the total reduces the weight of the relative error of the perturbation term, but when making the delta it does not contribute anymore.

Note that we have introduced statistical metrics Normalized Mean Bias (NMB) and Mean Bias (MB) to evaluate the agreement between FASST and TM5 with
Normalized Mean Bias = $\left(\overline{FASST - TM5}\right)/\overline{TM5}$ and
Mean Bias = $\left(\overline{FASST - TM5}\right)$

CHANGES TO MANUSCRIPT:
We included the following phrase in the re-written discussion in section 3.2 (P23 L13):

Contrary to $PM_{2.5}$, the NMB for the delta 6mDMA1 between two scenarios is higher than the NMB on absolute concentrations, with a low bias for the delta metric of -38% and -45% for Europe and North-America respectively, and a high bias of 35 to 46% in Asia. However, the MB on the delta is of the same order or lower than the absolute concentrations (Table 9). This is a consequence of the fixed background ozone in the absolute concentration reducing the weight of the anthropogenic fraction in the relative error.

30) 20.15-22: That's a reasonable comparison. I also wonder though what is the total number of estimated premature deaths associated with $PM_{2.5}$ and O_3, and how these numbers compare to those in the literature (from e.g. GBD), for present day conditions. This would help evaluate the accuracy of the absolute estimates in addition to estimates of changes.

REPLY: We have now included a table with some values from literature (both or $PM_{2.5}$ and O_3) in section 3.3 which is dedicated to a comparison with other published work, also illustrating the various assumptions that are involved making a direct comparison quite difficult.

CHANGES TO MANUSCRIPT: *Included a new section under section 3.3.4 Health impacts (P26 L24):*

Present-day health impacts

Table 14 gives an overview of recent global $PM_{2.5}$ health impact studies, together with FASST estimates for the year 2000 (RCP) and year 2010 (HTAP2 scenario). The studies differ in emission inventories and year evaluated, in applied methodologies to estimate $PM_{2.5}$ exposure, in model resolution, as well as in the choice of the exposure response functions, the value of the minimum exposure threshold, and mortality statistics. Studies excluding natural dust from the exposure are mostly applying the log-lin exposure response function and RR from Krewski et al. (2009), and estimate between 1.6 and 2.7 million annual premature mortalities from $PM_{2.5}$ in scenario years 2000 to 2004. FASST returns 2.1 and 2.5 million deaths using the GBD and log-lin exposure functions respectively. Studies including mineral dust are mostly applying the GBD integrated exposure-response functions and a non-zero threshold to avoid unrealistically high relative risk rates at high $PM_{2.5}$ levels in regions frequently exposed to dust. Depending on the choice of the exposure-response function and scenario year, FASST obtains 2.6 to 4.1 million global deaths, comparable with the range 1.7 to 4.2 million from previous studies.
Global ozone mortalities reported in Table 15 have been commonly based on the Jerrett et al. (2009) methodology, implemented in FASST. FASST obtains 197 thousand and 340 thousand deaths for RCP 2000 and HTAP2 2010 scenarios respectively, while the earlier studies find 380 to 470 thousand deaths in 2000, and 140 to 250 thousand in 2010 – 2015. Differences can be attributed to model chemical and meteorological processes, emission inventories, and the use of different sources for respiratory base mortality statistics.
Both for $PM_{2.5}$ and O_3, the difference between the different studies falls within the combined RR uncertainty and model variability range.

Table 14 Overview of previous studies on health impact of PM$_{2.5}$, together with FASST results for 2 different scenarios. Uncertainty ranges are as reported in the respective studies. The uncertainty range on FASST results includes the RR uncertainty only (Fig. S5.1 in the SI)

Reference	Year evaluated	Method	threshold	Exposure - response function	Global deaths (millions)
		Excluding mineral dust			
Fang et al., 2013	2000	CTM	no	K2009(a)	1.6 (1.2 – 1.9)
Silva et al., 2013	2000	CTM	no	K2009	2.1 (1.3 -3.0)
Anenberg et al., 2010	2000	CTM	5.8µg m^{-3}	K2009	2.7 (2.0 -3.4)
Evans et al., 2013	2004	SAT	5.8µg m^{-3}	K2009	2.7 (1.9 - 3.5)
Lelieveld et al., 2013	2005	CTM	no	K2009	2.2 (2.1 - 2.3)
FASST (RCP)	**2000**	**FASST**	**~7.3µg m^{-3}**	**K2009**	**2.5 (1.2 – 3.6)**
FASST (RCP)	**2000**	**FASST**	**~7.3µg m^{-3}**	**B2014(b)**	**2.1 (1.0 – 3.0)**
		Including mineral dust			
Silva et al., 2016	2000	ACCMIP CTM ensemble	~7.3µg m^{-3}	B2014	1.7 (1.3 – 2.1)
Evans et al. 2013	2004	SAT	5.8µg m^{-3}	K2009	4.3 (2.9 – 5.4)
Lelieveld et al., 2015	2010	CTM	~7.3µg m^{-3}	B2014	3.2 (1.5 - 4.6)
GBD2010 (Lim et al., 2012)	2010	Fused (FASST + SAT + ground based)	~7.3µg m^{-3}	B2014	3.2 (2.8 -3.6)
GBD2013 (Forouzanfar et al., 2015)	2013	Fused (FASST + SAT + ground based)	~7.3µg m^{-3}	B2014	2.9 (2.8 – 3.1)
GBD2015 (Cohen et al., 2017)	2015	Fused (FASST + SAT + ground based)	~4.1µg m^{-3}	B2014	4.2 (3.7 – 4.8)
FASST (RCP)	**2000**	**FASST**	**~7.3µg m^{-3}**	**K2009**	**3.6 (2.7 -4.5)**
FASST (RCP)	**2000**	**FASST**	**~7.3µg m^{-3}**	**B2014**	**2.6 (1.2 – 3.8)**
FASST (HTAP2)	**2010**	**FASST**	**~7.3µg m^{-3}**	**B2014**	**4.1 (2.0 - 5.9)**

(a) Krewski et al., 2009
(b) Burnett et al., 2014

Table 15 Overview of previous studies on long-term health impact of ozone, together with FASST results for 2 different scenarios

Ref	year	Method	threshold	Exposure-response function	Global deaths (thousands)
Anenberg et al., 2010	2000	CTM	33.3	J2009 (a)	470 (182 - 758)
Silva et al., 2013	2000	ACCMIP CTM ensemble	33.3	J2009	380 (117 -750)
Lelieveld et al., 2015	2010	CTM	~37.6	J2009	142 (90 -208)
GBD 2010 (Lim et al., 2012)	2010	FASST	~37.6	J2009	152 (52 – 270)
GBD 2013 (Forouzanfar et al., 2015)	2013	FASST	~37.6	J2009	217 (161 – 272)
GBF 2015 (Cohen et al., 2017)	2015	FASST	~37.6	J2009	254 (97 – 422)
FASST (RCP)	**2000**	**FASST**	**33.3**	**J2009**	**197 (66 – 315)**
FASST (HTAP2)	**2010**	**FASST**	**33.3**	**J2009**	**340 (116 – 544)**

(a) Jerrett et al., 2009

31) 20.25 - 21.7: I'm I incorrect in thinking that many of the pre-industrial to present IPCC RF's also include an 80% reduction in biomass burning sources? If so, this might further explain why the IPCC values are on the higher side. Also, IPCC estimates and those in Bond include RF of BC on snow, which I don't see as being accounted for in FASST.

REPLY:
We do not have the information on what reductions in biomass burning were assumed in IPCC models, but note that most recent studies point to smaller reductions, subject to large uncertainty. Large scale biomass burning is more prominent for OC emissions, than for BC. For instance, in the RCP2000 emission inventory, BC from large scale forest fires account for 15% of the total BC forcing, hence including BB does not make a large (absolute) difference on the already low BC forcing (from 0.15 to 0.17 W/m^2) and cannot account for the low bias. Other missing contributions could indeed be more relevant, like the BC mixing state and residence time, snow and ice albedo impacts and cloud interactions (see also our reply to comment 23).

32) 21.22: How do they know it's owing to different OH levels and lifetimes rather than to different emissions (line 21.14)?

REPLY: The reviewer makes a correct point. We cannot be certain about this statement. However, the Stevenson ACCMIP study was based on the same emissions database described by Lamarque et al. (2013) as used in this model study, which seems to point to differences in oxidation chemistry and resulting ozone production with respect to CO and NMVOC emissions. We also spotted an error in our data treatment and corrected the data in Table 10 which changes slightly the discussion.

CHANGES TO MANUSCRIPT *(P25 L6)*:
Table 10 compares the contribution of anthropogenic O_3 precursors CH_4, NO_x, NMVOC and CO to the O_3 and CH_4 radiative forcing with earlier work (Shindell et al., 2005, 2009; Stevenson et al., 2013). Except for NO_x which shows a large scatter across the studies, the FASST computed contributions to global O_3 and CH_4 forcing - using the same year 1850 to 2000 emission changes as in Stevenson et al. (2013) - are in good agreement with the model ensemble range in

the latter study. FASST NO_x forcing contributions are a factor 3 lower than in the Stevenson et al. study and more in line with Shindell et al. (2005, 2009) values (based on the period 1750 – 2000), however the latter obtain a NMVOC contribution to O_3 forcing which is a factor of 5 to 6 lower than the other estimates. Differences across the studies are likely due to differences in oxidation chemistry and lifetimes across models.

33) Section 3.3.2: The evaluation of global sector and species specific RF looks good. A key feature of FASST is regional specificity; could they also compare to some studies in the literature that have evaluation the RF of regionally specific emissions by species or sector?

REPLY: We note that FASST does not contain sector-specific SRs, hence global forcing efficiencies (expressed as $mW/m^2/Tg$) for a single FASST source region are valid for the aggregated contributions of the regional sectors.
The most relevant studies to compare aerosol global forcing responses to regional emissions are the HTAP1 exercise (Yu et al., 2013) and the similar multi-model HTAP2 study (Stjern et al., 2016). For the NH regions considered in these studies, our results correspond well (within 1 stdev) with older Yu et al. study (based on a single model, using similar emission and meteorological year as FASST base simulation), whereas the multi-model ensemble mean of Stjern et al. gives higher forcing efficiencies, although in the latter case the model variability is large, and our results stays within 2 stdv (95% confidence interval).

CHANGES TO MANUSCRIPT:
We have included an additional subsection under 3.3 (Comparison of TM5-FASST_v0 impact estimates with published studies) (P25 L15):

3.2.2 Regional forcing efficiencies by emitted component

Earlier work in the frame of HTAP1 (Fry et al., 2012; Yu et al., 2013) and HTAP2 (Stjern et al., 2016) evaluated regional forcing efficiencies for larger regions than the ones defined for FASST. For a comparison we aggregate the FASST forcing efficiencies (as listed in section S6.3 of the SI) by making an emission-weighted averages over Europe (EUR), North-America (NAM), South-Asia (SAS), East-Asia (EAS), Mediterranean and Middle East (MEA) and Russia, Belarus and Ukraine (RBU). Tables 11 (PM precursors) and 12 (NO_x and NMVOC) show the earlier studies along with the FASST results. The FASST forcing efficiencies for PM precursors confirm our earlier observation that FASST is particularly biased low for BC, in particular compared to Stjern et al. (2016), but further compares relatively well with the earlier work, in particular with Yu et al. (2013) which was based on a year 2001 baseline, similar to conditions of our base scenario. A similar observation is made for NMVOC for which FASST efficiencies agree well with the study by Fry et al. (2012). The forcing efficiency for ozone precursor NO_x has a high uncertainty. While for East-Asia, North-America and South-Asia the FASST result falls within 1 standard deviation of the HTAP1 model ensemble the FASST NO_x forcing efficiency for Europe shows a larger deviation. Without going into the details of the underlying mechanisms, ozone titration effects, which are better resolved with the higher TM5 model resolution, could be a contributing factor.

New tables 11 and 12:

Table 11. Regional-to-global direct radiative forcing efficiencies for $PM_{2.5}$ precursors (mW/m^2/Tg of annual emissions) for the lager source-receptor regions in earlier studies, and from FASST, aggregated to similar regional definitions. Values in brackets represent 1 standard deviation from the respective reported model ensembles.

		NAM	EUR	SAS	EAS	RUS	MEA
Stjern et al., 2016	BC	52 (±21)	55 (±22)	94 (±38)	55 (±16)	78 (±47)	202 (±323)
	POM	-8 (±6)	-7 (±4)	-10 (±6)	-5 (±3)	-2 (±5)	-18 (±7)
	SO4 (SO2)	-5 (±2)	-6 (±2)	-8 (±4)	-4 (±1)	-4 (±1)	-10 (±7)
Yu et al., 2013	BC	27 (±15)	37 (±19)	25 (±15)	28 (±20)		
	POM	-4 (±2)	-4 (±2)	-4 (±2)	-4 (±2)		
	SO4 (SO2)	-4 (±1)	-4 (±1)	-4 (±1)	-3 (±1)		
FASST (RCP2000)	BC	17	19	19	16	25	43
	POM	-6	-4	-6	-5	-4	-9
	SO4 (SO2)	-3	-3	-4	-2	-2	-7

Table 12. Regional-to-global direct radiative forcing efficiencies for O_3 precursors (mW/m^2/Tg of annual emissions) for the lager source-receptor regions in earlier work, and from FASST, aggregated to similar regional definitions, including feedbacks on CH4. Values in brackets represent reported 1 standard deviation from the model ensemble in the earlier work.

		EAS	EUR	NAM	SAS
Fry et al., 2010	NO_x	-0.22 (±0.6)	-1.20 (±0.5)	-0.48 (±0.6)	-1.70 (±2.2)
	NMVOC	0.42 (±0.2)	0.46 (±0.2)	0.42 (±0.2)	0.72 (±0.2)
FASST (RCP200)	NO_x	-0.44	-0.33	-0.35	-1.43
	NMVOC	0.60	0.57	0.61	0.74

34) 23.25: Does including this correction for changing mortality rates though lead to worse agreement between ACCMIP and FAST for $PM_{2.5}$ related deaths (Fig 15)?

REPLY:
It does not because according to the GBD methodology, respiratory mortality is not considered in the $PM_{2.5}$ related causes of death (which are: COPD, LC, IHD and Stroke), it contributes only to the O_3 health impact. The ACCMIP projections of PM-relevant base mortalities are much more in line with the ones used in FASST.

CHANGES TO MANUSCRIPT: *added the following phrase (P28 L25):*
Respiratory mortality is not considered as a cause of death for PM2.5, which explains why a similar disagreement is not observed in the $PM_{2.5}$ mortality trend in Fig. 17b.

35) Section 4: Good discussion. Some caveats about missing accurate treatment of SOA? Or carbonaceous aerosol aging? And possibly being a bit more clear about the limits of the emissions perturbations magnitudes that should be used with this tool (e.g., x2? x5? x10?).

REPLY:
Thank you for the positive feedback and the suggestions. We have extended some of the discussion making a wrap-up of the major caveats of the tool. Regarding the limits of the emission perturbations magnitudes, this depends on many parameters such as the region of

emission, on the emission ratio between various precursors, so it is not possible to set an overall validity range. We believe that the MIT and FLE scenarios explore the domain boundaries in which ‘reasonable’ emission changes for the next decades (until 2030) and that TM5-FASST behaves sufficiently well to be used as a screening tool to explore scenarios further out in the future.

CHANGES TO MANUSCRIPT

New discussion section 4.1 is – amongst other caveats - addressing the issues mentioned (P31 L16)

The current version of TM5-FASST is missing some source-receptor relations which may introduce a bias in estimated $PM_{2.5}$ and O_3 responses upon emission changes. The omission of secondary organic PM in TM5 is estimated to introduce a low bias in the base concentration of the order of 0.1 μg m^{-3} as global mean however with regional levels in Central Europe and China up to 1 μg m^{-3} in areas where levels of primary organic matter are reaching 20 μg m^{-3} (Farina et al., 2010) indicating a relatively low contribution of SOA to total PM2.5. O_3 formation from CO is included in the TM5 base simulations, but no SR matrices for the FASST source region definition are available. Based on the HTAP1 CO perturbation simulations with TM5, we estimate that a doubling of anthropogenic CO emissions contributes with 1 – 1.9 ppb in annual mean O_3 over Europe, 1.3 -1.9 ppb over North-America, 0.7-1.0 ppb over South Asia and 0.3 – 1.5 ppb over East-Asia. Development of CO-O_3 SRs is an important issue for the further development of the tool.

References

Amann, M., Bertok, I., Borken-Kleefeld, J., Cofala, J., Heyes, C., Höglund-Isaksson, L., Klimont, Z., Nguyen, B., Posch, M., Rafaj, P., Sandler, R., Schöpp, W., Wagner, F. and Winiwarter, W.: Cost-effective control of air quality and greenhouse gases in Europe: Modeling and policy applications, Environ. Model. Softw., 26(12), 1489–1501, doi:10.1016/j.envsoft.2011.07.012, 2011.

Andersson, C., Langner, J. and Bergström, R.: Interannual variation and trends in air pollution over Europe due to climate variability during 1958–2001 simulated with a regional CTM coupled to the ERA40 reanalysis, Tellus B, 59(1), 77–98, doi:10.1111/j.1600-0889.2006.00196.x, 2007.

Anenberg, S. C., Horowitz, L. W., Tong, D. Q. and West, J. J.: An estimate of the global burden of anthropogenic ozone and fine particulate matter on premature human mortality using atmospheric modeling, Environ. Health Perspect., 118(9), 1189–1195, doi:10.1289/ehp.0901220, 2010.

Anon: Estimates of global mortality attributable to particulate air pollution using satellite imagery, Environ. Res., 120, 33–42, doi:10.1016/j.envres.2012.08.005, 2013.

Bond, T. C., Doherty, S. J., Fahey, D. W., Forster, P. M., Berntsen, T., DeAngelo, B. J., Flanner, M. G., Ghan, S., Kärcher, B., Koch, D., Kinne, S., Kondo, Y., Quinn, P. K., Sarofim, M. C., Schultz, M. G., Schulz, M., Venkataraman, C., Zhang, H., Zhang, S., Bellouin, N., Guttikunda, S. K., Hopke, P. K., Jacobson, M. Z., Kaiser, J. W., Klimont, Z., Lohmann, U., Schwarz, J. P., Shindell, D., Storelvmo, T., Warren, S. G. and Zender, C. S.: Bounding the role of black carbon in the climate system: A scientific assessment, J. Geophys. Res. Atmospheres, 118(11), 5380–5552, doi:10.1002/jgrd.50171, 2013.

Burnett, R. T., Pope, C. A., III, Ezzati, M., Olives, C., Lim, S. S., Mehta, S., Shin, H. H., Singh, G., Hubbell, B., Brauer, M., Anderson, H. R., Smith, K. R., Balmes, J. R., Bruce, N. G., Kan, H., Laden, F., Prüss-Ustün, A., Turner, M. C., Gapstur, S. M., Diver, W. R. and Cohen, A.: An Integrated Risk Function for Estimating the Global Burden of Disease Attributable to Ambient Fine Particulate Matter Exposure, Environ. Health Perspect., doi:10.1289/ehp.1307049, 2014.

Christoudias, T., Pozzer, A. and Lelieveld, J.: Influence of the North Atlantic Oscillation on air pollution transport, Atmospheric Chem. Phys., 12(2), 869–877, doi:https://doi.org/10.5194/acp-12-869-2012, 2012.

Cohen, A. J., Brauer, M., Burnett, R., Anderson, H. R., Frostad, J., Estep, K., Balakrishnan, K., Brunekreef, B., Dandona, L., Dandona, R., Feigin, V., Freedman, G., Hubbell, B., Jobling, A., Kan, H., Knibbs, L., Liu, Y., Martin, R., Morawska, L., Pope, C. A., III, Shin, H., Straif, K., Shaddick, G., Thomas, M., van Dingenen, R., van Donkelaar, A., Vos, T., Murray, C. J. L. and Forouzanfar, M. H.: Estimates and 25-year trends of the global burden of disease attributable to ambient air pollution: an analysis of data from the Global Burden of Diseases Study 2015, The Lancet, 389(10082), 1907–1918, doi:10.1016/S0140-6736(17)30505-6, 2017.

Dentener, F., Keating, T., Akimoto, H., Pirrone, N., Dutchak, S., Zuber, A., Convention on Long-range Transboundary Air Pollution, United Nations and UNECE Task Force on Emission Inventories and Projections, Eds.: Hemispheric transport of air pollution 2010: prepared by the Task Force on Hemispheric Transport of Air Pollution acting within the framework of the Convention on Long-range Transboundary Air Pollution, United Nations, New York ; Geneva., 2010.

van Donkelaar, A., Martin, R. V., Brauer, M., Hsu, N. C., Kahn, R. A., Levy, R. C., Lyapustin, A., Sayer, A. M. and Winker, D. M.: Global Estimates of Fine Particulate Matter using a Combined Geophysical-Statistical Method with Information from Satellites, Models, and Monitors, Environ. Sci. Technol., 50(7), 3762–3772, doi:10.1021/acs.est.5b05833, 2016a.

van Donkelaar, A., Martin, R. V., Brauer, M., Hsu, N. C., Kahn, R. A., Levy, R. C., Lyapustin, A., Sayer, A. M. and Winker, D. M.: Global Estimates of Fine Particulate Matter using a Combined Geophysical-Statistical Method with Information from Satellites, Models, and Monitors, Environ. Sci. Technol., 50(7), 3762–3772, doi:10.1021/acs.est.5b05833, 2016b.

Fang, Y., Naik, V., Horowitz, L. W. and Mauzerall, D. L.: Air pollution and associated human mortality: the role of air pollutant emissions, climate change and methane concentration increases from the preindustrial period to present, Atmospheric Chem. Phys., 13(3), 1377–1394, doi:https://doi.org/10.5194/acp-13-1377-2013, 2013.

Farina, S. C., Adams, P. J. and Pandis, S. N.: Modeling global secondary organic aerosol formation and processing with the volatility basis set: Implications for anthropogenic secondary organic aerosol, J. Geophys. Res. Atmospheres, 115(D9), doi:10.1029/2009JD013046, 2010.

Fenech, S., Doherty, R. M., Heaviside, C., Vardoulakis, S., Macintyre, H. L. and O&apos;Connor, F. M.: The influence of model spatial resolution on simulated ozone and fine particulate matter for Europe:

implications for health impact assessments, Atmospheric Chem. Phys., 18(8), 5765–5784, doi:10.5194/acp-18-5765-2018, 2018.

Fiore, A. M., Jacob, D. J., Field, B. D., Streets, D. G., Fernandes, S. D. and Jang, C.: Linking ozone pollution and climate change: The case for controlling methane, Geophys. Res. Lett., 29(19), 25–1, 2002.

Fiore, A. M., West, J. J., Horowitz, L. W., Naik, V. and Schwarzkopf, M. D.: Characterizing the tropospheric ozone response to methane emission controls and the benefits to climate and air quality, J. Geophys. Res. Atmospheres, 113(8), doi:10.1029/2007JD009162, 2008.

Fiore, A. M., Dentener, F. J., Wild, O., Cuvelier, C., Schultz, M. G., Hess, P., Textor, C., Schulz, M., Doherty, R. M., Horowitz, L. W., MacKenzie, I. A., Sanderson, M. G., Shindell, D. T., Stevenson, D. S., Szopa, S., Van Dingenen, R., Zeng, G., Atherton, C., Bergmann, D., Bey, I., Carmichael, G., Collins, W. J., Duncan, B. N., Faluvegi, G., Folberth, G., Gauss, M., Gong, S., Hauglustaine, D., Holloway, T., Isaksen, I. S. A., Jacob, D. J., Jonson, J. E., Kaminski, J. W., Keating, T. J., Lupu, A., Marmer, E., Montanaro, V., Park, R. J., Pitari, G., Pringle, K. J., Pyle, J. A., Schroeder, S., Vivanco, M. G., Wind, P., Wojcik, G., Wu, S. and Zuber, A.: Multimodel estimates of intercontinental source-receptor relationships for ozone pollution, J. Geophys. Res. Atmospheres, 114(D4), doi:10.1029/2008JD010816, 2009.

Foley, K. M., Napelenok, S. L., Jang, C., Phillips, S., Hubbell, B. J. and Fulcher, C. M.: Two reduced form air quality modeling techniques for rapidly calculating pollutant mitigation potential across many sources, locations and precursor emission types, Atmos. Environ., 98, 283–289, doi:10.1016/j.atmosenv.2014.08.046, 2014.

Forouzanfar, M. H., Alexander, L., Anderson, H. R., Bachman, V. F., Biryukov, S., Brauer, M., Burnett, R., Casey, D., Coates, M. M., Cohen, A., Delwiche, K., Estep, K., Frostad, J. J., KC, A., Kyu, H. H., Moradi-Lakeh, M., Ng, M., Slepak, E. L., Thomas, B. A., Wagner, J., Aasvang, G. M., Abbafati, C., Ozgoren, A. A., Abd-Allah, F., Abera, S. F., Aboyans, V., Abraham, B., Abraham, J. P., Abubakar, I., Abu-Rmeileh, N. M. E., Aburto, T. C., Achoki, T., Adelekan, A., Adofo, K., Adou, A. K., Adsuar, J. C., Afshin, A., Agardh, E. E., Al Khabouri, M. J., Al Lami, F. H., Alam, S. S., Alasfoor, D., Albittar, M. I., Alegretti, M. A., Aleman, A. V., Alemu, Z. A., Alfonso-Cristancho, R., Alhabib, S., Ali, R., Ali, M. K., Alla, F., Allebeck, P., Allen, P. J., Alsharif, U., Alvarez, E., Alvis-Guzman, N., Amankwaa, A. A., Amare, A. T., Ameh, E. A., Ameli, O., Amini, H., Ammar, W., Anderson, B. O., Antonio, C. A. T., Anwari, P., Cunningham, S. A., Arnlöv, J., Arsenijevic, V. S. A., Artaman, A., Asghar, R. J., Assadi, R., Atkins, L. S., Atkinson, C., Avila, M. A., Awuah, B., Badawi, A., Bahit, M. C., Bakfalouni, T., Balakrishnan, K., Balalla, S., Balu, R. K., Banerjee, A., Barber, R. M., Barker-Collo, S. L., Barquera, S., Barregard, L., Barrero, L. H., Barrientos-Gutierrez, T., Basto-Abreu, A. C., Basu, A., Basu, S., Basulaiman, M. O., Ruvalcaba, C. B., Beardsley, J., Bedi, N., Bekele, T., Bell, M. L., Benjet, C., Bennett, D. A., et al.: Global, regional, and national comparative risk assessment of 79 behavioural, environmental and occupational, and metabolic risks or clusters of risks in 188 countries, 1990–2013: a systematic analysis for the Global Burden of Disease Study 2013, The Lancet, 386(10010), 2287–2323, doi:10.1016/S0140-6736(15)00128-2, 2015.

Fry, M. M., Naik, V., West, J. J., Schwarzkopf, M. D., Fiore, A. M., Collins, W. J., Dentener, F. J., Shindell, D. T., Atherton, C., Bergmann, D., Duncan, B. N., Hess, P., MacKenzie, I. A., Marmer, E., Schultz, M. G., Szopa, S., Wild, O. and Zeng, G.: The influence of ozone precursor emissions from four world regions on tropospheric composition and radiative climate forcing, J. Geophys. Res. Atmospheres, 117(7), doi:10.1029/2011JD017134, 2012.

Grewe, V., Dahlmann, K., Matthes, S. and Steinbrecht, W.: Attributing ozone to NOx emissions: Implications for climate mitigation measures, Atmos. Environ., 59, 102–107, doi:10.1016/j.atmosenv.2012.05.002, 2012.

Hu, J., Wang, P., Ying, Q., Zhang, H., Chen, J., Ge, X., Li, X., Jiang, J., Wang, S., Zhang, J., Zhao, Y. and Zhang, Y.: Modeling biogenic and anthropogenic secondary organic aerosol in China, Atmospheric Chem. Phys., 17(1), 77–92, doi:https://doi.org/10.5194/acp-17-77-2017, 2017.

Huang, Y., Wu, S., Dubey, M. and French, N. H. F.: Impact of aging mechanism on model simulated carbonaceous aerosols, Atmospheric Chem. Phys. Print, 12, 10.5194/acpd-12-28993–2012, doi:10.5194/acpd-12-28993-2012, 2012.

Huijnen, V., Williams, J., van Weele, M., van Noije, T., Krol, M., Dentener, F., Segers, A., Houweling, S., Peters, W., de Laat, J., Boersma, F., Bergamaschi, P., van Velthoven, P., Le Sager, P., Eskes, H., Alkemade, F., Scheele, R., Nédélec, P. and Pätz, H.-W.: The global chemistry transport model TM5: description and evaluation of the tropospheric chemistry version 3.0, Geosci. Model Dev., 3(2), 445–473, doi:10.5194/gmd-3-445-2010, 2010.

Jerrett, M., Burnett, R. T., Arden, P. I., Ito, K., Thurston, G., Krewski, D., Shi, Y., Calle, E. and Thun, M.: Long-term ozone exposure and mortality, N. Engl. J. Med., 360(11), 1085–1095, doi:10.1056/NEJMoa0803894, 2009.

Jin, X., Fiore, A. M., Murray, L. T., Valin, L. C., Lamsal, L. N., Duncan, B., Folkert, B., De, S., Abad, G. G., Chance, K. and Tonnesen, G. S.: Evaluating a Space-Based Indicator of Surface Ozone-NOx-VOC Sensitivity Over Midlatitude Source Regions and Application to Decadal Trends, J. Geophys. Res. Atmospheres, 122(19), 10439–10461, doi:10.1002/2017JD026720, 2017.

Kiesewetter, G., Borken-Kleefeld, J., Schöpp, W., Heyes, C., Thunis, P., Bessagnet, B., Terrenoire, E., Gsella, A. and Amann, M.: Modelling NO_2 concentrations at the street level in the GAINS integrated assessment model: projections under current legislation, Atmospheric Chem. Phys., 14(2), 813–829, doi:https://doi.org/10.5194/acp-14-813-2014, 2014.

Kiesewetter, G., Borken-Kleefeld, J., Schöpp, W., Heyes, C., Thunis, P., Bessagnet, B., Terrenoire, E., Fagerli, H., Nyiri, A. and Amann, M.: Modelling street level PM 10 concentrations across Europe: source apportionment and possible futures, Atmospheric Chem. Phys., 15(3), 1539–1553, 2015.

Kirschke, S., Bousquet, P., Ciais, P., Saunois, M., Canadell, J. G., Dlugokencky, E. J., Bergamaschi, P., Bergmann, D., Blake, D. R., Bruhwiler, L., Cameron-Smith, P., Castaldi, S., Chevallier, F., Feng, L., Fraser, A., Heimann, M., Hodson, E. L., Houweling, S., Josse, B., Fraser, P. J., Krummel, P. B., Lamarque, J.-F., Langenfelds, R. L., Le Quéré, C., Naik, V., O'Doherty, S., Palmer, P. I., Pison, I., Plummer, D., Poulter, B., Prinn, R. G., Rigby, M., Ringeval, B., Santini, M., Schmidt, M., Shindell, D. T., Simpson, I. J., Spahni, R., Steele, L. P., Strode, S. A., Sudo, K., Szopa, S., van der Werf, G. R., Voulgarakis, A., van Weele, M., Weiss, R. F., Williams, J. E. and Zeng, G.: Three decades of global methane sources and sinks, Nat. Geosci., 6(10), 813–823, doi:10.1038/ngeo1955, 2013.

Krewski, D., Jerrett, M., Burnett, R. T., Ma, R., Hughes, E. and Shi, Y.: Extended Follow-Up and Spatial Analysis of the American Cancer Society Study Linking Particulate Air Pollution and Mortality., Research Report, Health Effects Institute, Boston., 2009.

Lamarque, J.-F., Shindell, D. T., Josse, B., Young, P. J., Cionni, I., Eyring, V., Bergmann, D., Cameron-Smith, P., Collins, W. J., Doherty, R., Dalsoren, S., Faluvegi, G., Folberth, G., Ghan, S. J., Horowitz, L. W., Lee, Y. H., MacKenzie, I. A., Nagashima, T., Naik, V., Plummer, D., Righi, M., Rumbold, S. T., Schulz, M., Skeie, R. B., Stevenson, D. S., Strode, S., Sudo, K., Szopa, S., Voulgarakis, A. and Zeng, G.: The Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP): overview and description of models, simulations and climate diagnostics, Geosci Model Dev, 6(1), 179–206, doi:10.5194/gmd-6-179-2013, 2013.

Lelieveld, J., Barlas, C., Giannadaki, D. and Pozzer, A.: Model calculated global, regional and megacity premature mortality due to air pollution, Atmospheric Chem. Phys., 13(14), 7023–7037, doi:https://doi.org/10.5194/acp-13-7023-2013, 2013.

Li, J., Yang, W., Wang, Z., Chen, H., Hu, B., Li, J., Sun, Y. and Huang, Y.: A modeling study of source–receptor relationships in atmospheric particulate matter over Northeast Asia, Atmos. Environ., 91, 40–51, doi:10.1016/j.atmosenv.2014.03.027, 2014.

Li, Q., Jacob, D. J., Bey, I., Palmer, P. I., Duncan, B. N., Field, B. D., Martin, R. V., Fiore, A. M., Yantosca, R. M., Parrish, D. D., Simmonds, P. G. and Oltmans, S. J.: Transatlantic transport of pollution and its effects on surface ozone in Europe and North America, J. Geophys. Res. Atmospheres, 107(D13), ACH 4-1-ACH 4-21, doi:10.1029/2001JD001422, 2002.

Li, Y., Henze, D. K., Jack, D. and Kinney, P. L.: The influence of air quality model resolution on health impact assessment for fine particulate matter and its components, Air Qual. Atmosphere Health, 9(1), 51–68, doi:10.1007/s11869-015-0321-z, 2016.

Lim, S. S., Vos, T., Flaxman, A. D., Danaei, G., Shibuya, K., Adair-Rohani, H., Amann, M., Anderson, H. R., Andrews, K. G., Aryee, M., Atkinson, C., Bacchus, L. J., Bahalim, A. N., Balakrishnan, K., Balmes, J., Barker-Collo, S., Baxter, A., Bell, M. L., Blore, J. D., Blyth, F., Bonner, C., Borges, G., Bourne, R., Boussinesq, M., Brauer, M., Brooks, P., Bruce, N. G., Brunekreef, B., Bryan-Hancock, C., Bucello, C., Buchbinder, R., Bull, F., Burnett, R. T., Byers, T. E., Calabria, B., Carapetis, J., Carnahan, E., Chafe, Z., Charlson, F., Chen, H., Chen, J. S., Cheng, A. T.-A., Child, J. C., Cohen, A., Colson, K. E., Cowie, B. C., Darby, S., Darling, S., Davis, A., Degenhardt, L., Dentener, F., Des Jarlais, D. C., Devries, K., Dherani, M., Ding, E. L., Dorsey, E. R., Driscoll, T., Edmond, K., Ali, S. E., Engell, R. E., Erwin, P. J., Fahimi, S., Falder, G., Farzadfar, F., Ferrari, A., Finucane, M. M., Flaxman, S., Fowkes, F. G. R., Freedman, G., Freeman, M. K., Gakidou, E., Ghosh, S., Giovannucci, E., Gmel, G., Graham, K., Grainger, R., Grant, B., Gunnell, D., Gutierrez, H. R., Hall, W., Hoek, H. W., Hogan, A., Hosgood III, H. D., Hoy, D., Hu, H., Hubbell, B. J., Hutchings, S. J., Ibeanusi, S. E., Jacklyn, G. L., Jasrasaria, R., Jonas, J. B., Kan, H., Kanis, J. A., Kassebaum, N., Kawakami, N., Khang, Y.-H., Khatibzadeh, S., Khoo, J.-P., Kok, C., et al.: A comparative risk assessment of burden of disease and injury attributable to 67 risk factors and risk factor clusters in 21 regions, 1990-2010: A systematic analysis for the Global Burden of Disease Study 2010, The Lancet, 380(9859), 2224–2260, doi:10.1016/S0140-6736(12)61766-8, 2012.

Liu, X., Penner, J. E., Das, B., Bergmann, D., Rodriguez, J. M., Strahan, S., Wang, M. and Feng, Y.: Uncertainties in global aerosol simulations: Assessment using three meteorological data sets, J. Geophys. Res., 112(D11), doi:10.1029/2006JD008216, 2007.

Liu, Y., Hong, Y., Fan, Q., Wang, X., Chan, P., Chen, X., Lai, A., Wang, M. and Chen, X.: Source-receptor relationships for PM2.5 during typical pollution episodes in the Pearl River Delta city cluster, China, Sci. Total Environ., 596–597, 194–206, doi:10.1016/j.scitotenv.2017.03.255, 2017.

Maione, M., Fowler, D., Monks, P. S., Reis, S., Rudich, Y., Williams, M. L. and Fuzzi, S.: Air quality and climate change: Designing new win-win policies for Europe, Environ. Sci. Policy, 65, 48–57, doi:10.1016/j.envsci.2016.03.011, 2016.

Ming, Y. and Russell, L. M.: Predicted hygroscopic growth of sea salt aerosol, J. Geophys. Res. Atmospheres, 106(D22), 28259–28274, 2001.

Myhre, G., Fuglestvedt, J. S., Berntsen, T. K. and Lund, M. T.: Mitigation of short-lived heating components may lead to unwanted long-term consequences, Atmos. Environ., 45(33), 6103–6106, doi:10.1016/j.atmosenv.2011.08.009, 2011.

Pausata, F. S. R., Pozzoli, L., Dingenen, R. V., Vignati, E., Cavalli, F. and Dentener, F. J.: Impacts of changes in North Atlantic atmospheric circulation on particulate matter and human health in Europe, Geophys. Res. Lett., 40(15), 4074–4080, doi:10.1002/grl.50720, 2013.

Pope, R. J., Chipperfield, M. P., Arnold, S. R., Glatthor, N., Feng, W., Dhomse, S. S., Kerridge, B. J., Latter, B. G. and Siddans, R.: Influence of the wintertime North Atlantic Oscillation on European tropospheric composition: an observational and modelling study, Atmospheric Chem. Phys., 18(11), 8389–8408, doi:10.5194/acp-18-8389-2018, 2018.

Porter, P. S., Rao, S. T., Hogrefe, C. and Mathur, R.: A reduced form model for ozone based on two decades of CMAQ simulations for the continental United States, Atmospheric Pollut. Res., 8(2), 275–284, doi:10.1016/j.apr.2016.09.005, 2017.

Prather, M., Ehhalt, D., Dentener, F., Derwent, R., Dlugokencky, E., Holland, E., Isaksen, I., Katima, J., Kirchhoff, V., Matson, P., Midgley, P., Wang, M., Berntsen, T., Bey, I., Brasseur, G., Buja, L., Pitari, G. and Et, A.: Chapter 4: Atmospheric Chemistry and Greenhouse Gases, Cambridge University Press. [online] Available from: https://ricerca.univaq.it/handle/11697/24359#.WItxbn0XGao (Accessed 27 January 2017), 2001.

Punger, E. M. and West, J. J.: The effect of grid resolution on estimates of the burden of ozone and fine particulate matter on premature mortality in the United States, Air Qual. Atmosphere Health, 6(3), doi:10.1007/s11869-013-0197-8, 2013.

Shindell, D., Kuylenstierna, J., Vignati, E., van Dingenen, R., Amann, M., Klimont, Z., Anenberg, S., Muller, N., Janssens-Maenhout, G., Raes, F., Schwartz, J., Faluvegi, G., Pozzoli, L., Kupiainen, K., Hoglund-Isaksson, L., Emberson, L., Streets, D., Ramanathan, V., Hicks, K., Oanh, N., Milly, G., Williams, M., Demkine, V. and Fowler, D.: Simultaneously Mitigating Near-Term Climate Change and Improving Human Health and Food Security, Science, 335(6065), 183–189, doi:10.1126/science.1210026, 2012.

Shindell, D. T., Faluvegi, G., Bell, N. and Schmidt, G. A.: An emissions-based view of climate forcing by methane and tropospheric ozone: EMISSIONS-BASED CLIMATE FORCING, Geophys. Res. Lett., 32(4), n/a-n/a, doi:10.1029/2004GL021900, 2005.

Shindell, D. T., Faluvegi, G., Koch, D. M., Schmidt, G. A., Unger, N. and Bauer, S. E.: Improved Attribution of Climate Forcing to Emissions, Science, 326(5953), 716–718, doi:10.1126/science.1174760, 2009.

Silva, R. A., West, J. J., Zhang, Y., Anenberg, S. C., Lamarque, J.-F., Shindell, D. T., Collins, W. J., Dalsoren, S., Faluvegi, G., Folberth, G., Horowitz, L. W., Tatsuya Nagashima, Naik, V., Rumbold, S., Skeie, R., Sudo, K., Takemura, T., Daniel Bergmann, Cameron-Smith, P., Cionni, I., Doherty, R. M., Eyring, V., Josse, B., MacKenzie, I. A., Plummer, D., Righi, M., Stevenson, D. S., Strode, S., Szopa, S. and Zeng, G.: Global premature mortality due to anthropogenic outdoor air pollution and the contribution of past climate change, Environ. Res. Lett., 8(3), 034005, doi:10.1088/1748-9326/8/3/034005, 2013.

Silva, R. A., West, J. J., Lamarque, J. F., Shindell, D. T., Collins, W. J., Dalsoren, S., Faluvegi, G., Folberth, G., Horowitz, L. W., Nagashima, T., Naik, V., Rumbold, S. T., Sudo, K., Takemura, T., Bergmann, D., Cameron-Smith, P., Cionni, I., Doherty, R. M., Eyring, V., Josse, B., MacKenzie, I. A., Plummer, D., Righi, M., Stevenson, D. S., Strode, S., Szopa, S. and Zengast, G.: The effect of future ambient air pollution on human premature mortality to 2100 using output from the ACCMIP model ensemble, Atmospheric Chem. Phys., 16(15), 9847–9862, doi:10.5194/acp-16-9847-2016, 2016.

Stevenson, D. S., Young, P. J., Naik, V., Lamarque, J.-F., Shindell, D. T., Voulgarakis, A., Skeie, R. B., Dalsoren, S. B., Myhre, G., Berntsen, T. K., Folberth, G. A., Rumbold, S. T., Collins, W. J., MacKenzie, I. A., Doherty, R. M., Zeng, G., Van, N., Strunk, A., Bergmann, D., Cameron-Smith, P., Plummer, D. A., Strode, S. A., Horowitz, L., Lee, Y. H., Szopa, S., Sudo, K., Nagashima, T., Josse, B., Cionni, I., Righi, M., Eyring, V., Conley, A., Bowman, K. W., Wild, O. and Archibald, A.: Tropospheric ozone changes, radiative forcing and attribution to emissions in the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP), Atmospheric Chem. Phys., 13(6), 3063–3085, doi:10.5194/acp-13-3063-2013, 2013.

Stier, P., Feichter, J., Kinne, S., Kloster, S., Vignati, E., Wilson, J., Ganzeveld, L., Tegen, I., Werner, M., Balkanski, Y., Schulz, M., Boucher, O., Minikin, A. and Petzold, A.: The aerosol-climate model ECHAM5-HAM, Atmospheric Chem. Phys., 5(4), 1125–1156, doi:https://doi.org/10.5194/acp-5-1125-2005, 2005.

Stjern, C. W., Samset, B. H., Myhre, G., Bian, H., Chin, M., Davila, Y., Dentener, F., Emmons, L., Flemming, J., Haslerud, A. S., Henze, D., Jonson, J. E., Kucsera, T., Lund, M. T., Schulz, M., Sudo, K., Takemura, T. and Tilmes, S.: Global and regional radiative forcing from 20 % reductions in BC, OC and SO4 - An HTAP2 multi-model study, Atmospheric Chem. Phys., 16(21), 13579–13599, doi:10.5194/acp-16-13579-2016, 2016.

Tang, I. N.: Chemical and size effects of hygroscopic aerosols on light scattering coefficients, J. Geophys. Res. Atmospheres, 101(14), 19245–19250, 1996.

Turnock, S., Wild, O., Dentener, F., Davila, Y., Emmons, L., Flemming, J., Folberth, G., Henze, D., Jonson, J., Keating, T., Kengo, S., Lin, M., Lund, M., Tilmes, S. and O’Connor, F.: The Impact of Future Emission Policies on Tropospheric Ozone using a Parameterised Approach, Atmospheric Chem. Phys. Discuss., 1–41, doi:https://doi.org/10.5194/acp-2017-1220, 2018.

Van Dingenen, R., Dentener, F. J., Raes, F., Krol, M. C., Emberson, L. and Cofala, J.: The global impact of ozone on agricultural crop yields under current and future air quality legislation, Atmos. Environ., 43(3), 604–618, doi:10.1016/j.atmosenv.2008.10.033, 2009.

West, J. J., Fiore, A. M., Naik, V., Horowitz, L. W., Schwarzkopf, M. D. and Mauzerall, D. L.: Ozone air quality and radiative forcing consequences of changes in ozone precursor emissions, Geophys. Res. Lett., 34(6), doi:10.1029/2006GL029173, 2007.

Wild, O. and Prather, M. J.: Excitation of the primary tropospheric chemical mode in a global three-dimensional model, J. Geophys. Res. Atmospheres, 105(D20), 24647–24660, 2000.

Wild, O., Fiore, A. M., Shindell, D. T., Doherty, R. M., Collins, W. J., Dentener, F. J., Schultz, M. G., Gong, S., MacKenzie, I. A., Zeng, G. and others: Modelling future changes in surface ozone: a parameterized approach, Atmospheric Chem. Phys., 12(4), 2037–2054, 2012.

Wu, S., Duncan, B. N., Jacob, D. J., Fiore, A. M. and Wild, O.: Chemical nonlinearities in relating intercontinental ozone pollution to anthropogenic emissions, Geophys. Res. Lett., 36(5), doi:10.1029/2008GL036607, 2009.

Yu, H., Chin, M., West, J. J., Atherton, C. S., Bellouin, N., Bergmann, D., Bey, I., Bian, H., Diehl, T., Forberth, G., Hess, P., Schulz, M., Shindell, D., Takemura, T. and Tan, Q.: A multimodel assessment of the influence of regional anthropogenic emission reductions on aerosol direct radiative forcing and the role of intercontinental transport, J. Geophys. Res. Atmospheres, 118(2), 700–720, doi:10.1029/2012JD018148, 2013.

Zhang, L., Liu, L., Zhao, Y., Gong, S., Zhang, X., Henze, D. K., Capps, S. L., Fu, T.-M., Zhang, Q. and Wang, Y.: Source attribution of particulate matter pollution over North China with the adjoint method, Environ. Res. Lett., 10(8), 084011, 2015.