

Interactive comment on “Twenty-five years of continuous sulphur dioxide emission reduction in Europe” by V. Vestreng et al.

V. Vestreng et al.

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Response to referee, J. Olivier We would like to thank the referee for his contribution to increase the scientific value and readability of our paper. We highly appreciate his characteristic of our paper as important, and that he acknowledge our efforts to document not only the emission estimates but also their quality, validation and verification.

The referee comments are addressed one by one below, except for his comments on the NEC targets which are covered together. There were some discrepancies between the page numbering in the manuscript, and the one used by the referee. We do think that we have located the comments to the appropriate places in the manuscript, but apologize beforehand if our interpretation is not correct. We take his references to p. 52xx as references to 51xx and have added page numbers to comments where we anticipated these to be missing in the referees response.

Specific comments:

p. 5102, line 1-2: I miss the NEC targets in a column in Table 2.

p. 5104, line 16-17: Why not also compared to NEC targets?

p. 5128, line 26: I miss here . I also miss here a remark on the more strict NEC targets for EU countries.

Response: We have deliberately chosen not to include more specifically the targets as outlined in the NEC Directive, because the emission reporting requirements under the NEC Directive (NECD) and the Convention on LRTAP differs. In addition, a complete set of SO₂ NEC emission totals is only available for the EU-15 from 2001-2004, thus no (long-term) NEC trends are available. We acknowledge that it is certainly not obvious to all readers why we have not included also the NEC targets (and emission estimates) explicitly in our paper, and we will add a sentence on this in the final version of the manuscript;

p. 5104, line 17: (Gothenburg Protocol). Emission targets in the NEC Directive are not included, as the emission reporting requirements, hence the national total emissions reported, differs on several points between the LRTAP Convention and the NEC Directive. In addition, final NEC emission data is only complete for the EU-15 total emissions for five years (2000-2004); hence analysis of long-term trends which is the focus in this paper cannot be undertaken.

p. 5102, line 9: from biofuel use, forest fires ..

Response: This paragraph concerns natural emissions of sulphur; we therefore hesitate to include biofuel use here.

p. 5102, line 16: pls. explain the term critical loads, which I expect is not familiar for most readers.

Response: Accepted. In the final version of the document we will insert a footnote 1

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and accompanying references on critical loads¹.

Footnote 1. The basic idea of the critical load is to balance the deposition rate to an ecosystem with its long-term capability to buffer the input or to remove it without harmful effects inside or outside the system (Hettelingh et al., 2001; UBA, 2004)

References:

Hettelingh, J.-P., Posh, M. and De Smet, P.A.M.: Multi-Effect critical loads used in multi-pollutant reduction agreements in Europe. *Water, Air and Soil Pollution* 130, 1133-1138, 2001 UBA (Ed): Manual on methodologies and criteria for modelling and mapping critical loads and levels, and air pollution effects, risks and trends. German Federal Environmental Agency, Berlin, Germany, 2004. Continuous updates available from <http://www.icpmapping.org>

p. 5103, line 10: than before.: recommend to also mention the old limits.

Response: Accepted. This will be included in the final version of the manuscript as: p. 5103, line 8-10: Hence the limit values of 20 μgm^{-3} for 24 hours average exposure and 500 μgm^{-3} for a 10-minute average are much more stringent than in the 2000 revision of the Guidelines where the limit was 125 μgm^{-3} as a 24 hours average (WHO, 2005).

p. 5104, line 2: I do not understand +-5-6%. Is it +-5% or +-6%??

Response: This is a typo, and will be corrected to +-5% in the final version of the manuscript.

p. 5105, line 5-7: The source categories that are included in the inventory are not well defined and described. Please mention the number and level of detail of source categories reported, e.g. total industrial combustion or per major subsector, e.g. iron and steel, chemical industry.

Response: Accepted. We will include the following paragraph and correction in the final version of the manuscript;

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p. 5105, line 7: EMEP programme. The emission data are reported in the Nomenclature For Reporting (NFR) source categories. There are 102 NFR categories in the reporting templates (<http://www.emep.int/emis2006/reportinginstructions.html>), including both detailed categories (e.g. Residential plants, Passenger cars and Iron and Steel, in addition to Public Electricity and Heat Production) as well as the associated aggregated levels (e.g. Residential, Road Transport and Manufacturing Industries and Construction) to facilitate reporting under the Convention also for Parties with less resources available for emission estimation and reporting. Reporting according to NFR mostly applies for the 1990 and onwards emission data. The 1980s are still dominated by emission data in the eleven SNAP (Selected Nomenclature for Air Pollutants) source sectors as defined in the EMEP/CORINAIR Guidebook (<http://reports.eea.europa.eu/EMEPCORINAIR4/en>), i.e. less detailed information is available for this time period.

p. 5105 line 10: should then read: National experts are requested to estimate their national emissions according to the EMEP/CORINAIR Emission Inventory Guidebook. Delete: (<http://reports.eea.europa.eu/EMEPCORINAIR4/en>). p. 5106, line 7: should then read: For the scope of this study, the emission sector data is presented according to SNAP source sectors.

p. 5105, line 9-10: The methodologies used for the whole time series are not very specific. Since when are estimates made cf. the Guidebook? What are the main differences with previous methodologies/guidebooks.

Response: It is acknowledged that the EMEP inventory differs from many other inventories in that it is compiled from a multitude of country specific national inventories, and not estimated by one institutional emission module based on international statistics, most relevant emission factors per area/country/process (dependant on availability), and if available techno-economic parameters. In order to assure methodological comparability between the different national inventories, EMEP, through its Task Force on Emission Inventories and Projections (TFEIP) has developed the EMEP/CORINAIR

Guidebook (GB) for emission estimation. The methodologies for emission estimation described in the GB have not changed considerably over the last 25 years and parties are applying the GB as we state at p. 5109, line 9. The GB has however a three Tier approach for emission estimation (IPCC, 2006). By moving from a lower to a higher Tier, more specific emission factors, more detailed activity information, specific abatement strategies and other relevant technical information is required. We have already addressed the request for recalculations of the whole time series whenever updates such as moving to a higher Tier, implementation of more specific EFs and activity data and correction of detected errors in input parameters to emission calculations (e.g. on p. 5107, line 2-5 and on p. 5109, line 15-19). Due to limited resources, it is sometimes the case that only later years and or round years (1990, 1995, 2000), are calculated applying a higher Tier. This will lead to inconsistencies in the timeseries as described on p. 5019, line 17. In order to clarify the methodological framework utterly we will add one paragraph in the final version of the manuscript:

p. 5105, line 11: After the revised sentence National experts are requested to estimate their national emissions according to the EMEP/CORINAIR Emission Inventory Guidebook insert

The Guidebook offers a three Tier approach for emission estimation (<http://www.ipcc-nggip.iges.or.jp/public/2006gl> for a definition of Tiers). By moving from a lower to a higher Tier, more specific emission factors, more detailed activity information, specific abatement strategies and other relevant technical information is required. The Tier approach allows all Parties to apply the Guidebook for their emission estimation irrespectively of resources and or detail of information available for emission estimation, as emissions can be estimated on different levels of complexity.

p. 5105, line 11-12: Are these also documented for the older emissions (e.g. for 1980)?

Response: It depends on the level of recalculation. Most Parties have reported their 1980s emission data according to SNAP, while in later years all Parties report according

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to NFR format. The documentation of country specific methodologies differ between the countries providing Informative Inventory Reports (50% of those reporting emission data), and information about 1980s emission data are generally only available for those countries which have recalculated their whole time series according to NFR.

We will add more information to clarify this issue in the final version of the manuscript: p.5105, line 12:...separately. This documentation is rather scarce for the 1980s, as it is only the past few years that the requirements for national emission data have become more transparent and guidelines on emission reporting, requesting also Informative Inventory Reports (IIRs), have become available (UNECE, 2003). In addition, new routines and standards for validating emission data have recently been adopted (UNECE, 2005).

p. 5109, line 17: However, sometimes only the latest year or data back to 1990 are recalculated by applying best available methodologies and emission factors, while the remaining part of the timeseries consists of data reported according to SNAP source categories.

p. 5109, line 23. The fact that an inventory consists of reported data in different formats does not necessarily mean that the data are not consistent according to the emission data review. p. 5107, line 23-24: Does replacement also take place if that would result again in an outlier?

Response: By default no. We will describe more carefully the methodology for replacements to clarify this also in relation to the referees comment below p. 5136: Table 1.

We will add the a sentence to the final version of the manuscript p. 5107, line 24: As mentioned also in section 2, replacements might simply consist of linear interpolation between two adjacent years. Whenever outliers are identified several places in the inventory, it is necessary to replace the whole timeseries by independent estimates, as the latter very rarely will be consistent with the emission data provided by the country

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itself.

p. 5109, line 6: Why tentatively? It IS an outlier. You probably mean identified as outlier, and thus possibly an error, which could however be well explained by national or source-specific circumstances.

Response: Accepted. The sentence will be corrected accordingly in the revise the final version of the manuscript to read: p. 5109, line 6: Results which fall outside the empirical ranges of averages are identified as outliers by the review team of experts, thus possibly an error, which could however be well explained by national and source-specific circumstances.

p. 5111, line 4-5: I would recommend to add the % of emissions and % of land area covered by these 14 countries.

Response: Accepted. We apologize for a typo in this sentence. The correct number of countries which report gridded sector data of sufficient quality to be included in the model calculations is 12 and not 14; Austria, Denmark, Finland, France, Germany, Lithuania, Netherlands, Norway, Spain, Sweden, Switzerland and United Kingdom. Emissions from these countries to the total anthropogenic land based emissions of SO₂ amount to 3566 Gg or 24% in 2004 (Table 1). These countries cover 25% of the total area covered by countries included in the EMEP domain of calculation.

We will correct the final version of the manuscript p. 5111, line 3-7 to: Gridded sector data is requested in 5-yearly intervals from 1990 onwards, but only twelve Parties to the LRTAP Convention have reported gridded sector data of any vintage in the 50x50km² EMEP grid by 2006 (<http://www.emep.int/grid/>). These countries represent 24% of the emissions and 25% of the area covered by the Parties listed in Table 1. EMEP is thus required to account for the spatial distribution of emissions for a large part of Europe by deriving its own methods.

p. 5112, line 1: Bouwman, 1997 should be et al., but more important that paper does

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not provide trends so I do not see the relevance of citing it in this context.

Response: The reference here to Bouwman et al., 1997 will not appear in the final version of the manuscript, hence has to be removed from the Reference list p. 5129, line 23.

p. 5113, line 13-14: increasingly overestimated over the years and indicate .. trend .. is reasonable are rather vague expressions. I recommend to provide some quantification for these statements.

Response: The paragraph will be corrected as follows in the final version of the manuscript. p. 5113, line 13-17: Sulphur dioxide was increasingly overestimated over the years from 122% (modelled concentrations are on average more than twice as large as observed) in 1985 to 349% in 2000. Although the modelled and measured decreases for sulphate 1985-2000 correspond within a few percent, indicating that the trend in the emission data is reasonable, it is difficult to conclude more specifically on the quantity of the emission trend, as the trends in the primary component SO₂ (which are closely related to the emission changes) are not very well reproduced in their model simulations

p. 5113, line 24: somewhat too low. Ibidem. and p. 5113, line 27-30: better reproduced .. indicating .. trend .. correspond to .. conc.. Ibidem.

Response: We agree that some more clarification and quantification is desirable in this paragraph, and will correct the final version of the document as follows:

p. 5113, line 23-30: Similar to Berglen et al. (2006), the decrease in SO₂ concentrations between 1980 and 2000 was too low compared to the decrease in observations. Whilst modelled SO₂ concentrations in 1980 were in good agreement with the observations (on average overestimated by 4%), the model simulations overestimated SO₂ by 39% in 2000. In the EMEP Unified model, a parameterisation of the so-called co-deposition of NH₃ and SO₂ is implemented in the dry deposition module (Simpson et

al, 2003). Fagerli et al. (in preparation) demonstrate that if this effect is not taken into account, the overestimation of SO₂ around 2000 is even larger (around 80%), whilst it has little effect around 1980 when SO₂ emissions peaked. Thus, a part of the increasing overestimation of SO₂ as calculated by Berglen et al. (2006) can be explained by the rather simple dry deposition scheme in their model. It is not clear why the EMEP model still overestimate SO₂ for recent years. However, the major part of the reduction in the SO₂ concentrations are captured by the model simulations, hence the trend in the EMEP SO₂ emission inventory does correspond to the observed decreases in SO₂ concentrations.

p. 5114: Ch. 4: Would Chapter 4 not be more logically placed before Ch. 3? Then Ch. 6 with discussion follows more logically the text on validation.

Response: We understand well this proposal, as we also did consider the order of chapters proposed by the referee. The reason why we finally decided to structure the paper the way we did is that we first want to build the confidence in the emission data included in the analysis both with respect to bottom-up (Ch2) and top-down (Ch3) validation, before we present the results (Ch4). In fact we emphasis the background for this decision in the introduction where we write: p. 5014, line 25 etc.: The emission improvement program under EMEP has increased the transparency and confidence in official submission and is a main reason to support for the first time the publication of the EMEP trends. Hence, we would like to keep the order of chapters to remain unchanged.

p. 5117, line 20: ..(FGD), mainly in power generation, ..

Response: Accepted. The correction will be made in the final version of the manuscript.

p. 5117, line 21: to gas or to nuclear power.

Response: Accepted. The correction will be made in the final version of the manuscript.

p. 5120, Section 4.2: As mentioned before, I miss here a short discussion of the targets

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of the NEC Directive.

Response: Please see Response above on this topic.

p. 5121, line 26: For others, not familiar with conventions in uncertainties of emissions, please add that the uncertainties mentioned here are estimates for the 95% confidence interval (i.e. reflect 2 sigma of a normal distribution).

Response: Accepted. We will change the final version of the manuscript p. 5121, line 26 to: The overall uncertainty for all GHG was shown to be 4-8% measured as 95% confidence intervals.

p. 5123, line 4: amount above +/-20%. Pls. provide a more approximate value or range.

Response: Accepted. We will revise the sentence in the final version of the manuscript to read: p. 5123, line 2: According to Schöpp et al. (2005), data for some Central and Eastern European countries are more uncertain than for the EU-15 countries, but the uncertainty do not exceed +/-23% for any country.

p. 5123, line 5-6: EMEP inventory total .. between 3% and 25% for individual countries after 1990. This is unclear: the first part suggests that the 3 to 25% refers to the EMEP total area of all countries, whereas the latter refers to specific countries. Do you mean that is the uncertainty estimate for the total EMEP area now estimated at 13 (+- 10)%?

Response: Accepted. We will revise the final version of the manuscript by substituting: P 5123, line 4: Since inclusion of non-official emission estimates is typically required for the latter countries, the uncertainty in the EMEP inventory total emissions is indicated to be between 3% and 25% for individual countries after 1990 with We therefore conclude that the uncertainty in the post 1990 emission estimates for individual countries included in the EMEP inventory lies between 3% and 25%, implying that the uncertainty in the EMEP inventory as such is less.

p. 5124, line 20-21: The 1990 SO₂ emission factors provided by Berdowski are country-specific and based on the sulphur contents of different fuels and metal ores

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smelted and country-specific sulphur recovery by smelters, refineries and FGD in power generation.

Response: We will add a sentence in the final version of the manuscript: P.4124, line 24, before Lefohn et.: The emission factors provided by Berdowski are country-specific and based on the sulphur contents of different fuels and metal ores smelted and country-specific sulphur recovery by smelters, refineries and FGD in power generation (J.Olivier pers. com. 2007).

Note: We could not find another appropriate reference for the information in this sentence although we have searched for it. There is a reference in van Aardenne et al. (2001) p. 912, Table 2, footnote a, to Olivier et al. (2000). This reference does not appear in the reference list, so we do not know if this is an appropriate reference for the information proposed by the referee. We regard this as unlikely as it does not appear in Olivier et al. (2002), p. 164 of this thesis. If such a reference exists, we will include it in the final version of the document.

p. 5126, line 22-23: .. implementation of measures and to economic recession.. Pls. add the fraction or % of each. I miss here the importance of changes in the fuel mix, notably from coal to oil and gas, which are often not due to policy regulation but e.g. to prices. p. 5127, line 2-14. I miss here a discussion of the importance of changes in the fuel mix, notably from coal to oil and gas.

Response: Based on the two comments above, we will revise the paragraph p. 5127, line 2-14 in the final version of the manuscript to read: Our analysis shows that different emission reduction drivers have different level of importance depending on which period and which part of Europe we are looking at. While the Eastern European changes in fuel consumption is directly reflected in the sulphur emissions during the first reduction regime, the Western European emissions are already decoupled from the fuel consumption thanks also to policy regulations already from the early 1980s, leading to implementation of advanced emission control technologies and changes in the fuel mix

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notably from coal to oil and gas. During the second reduction regime, the economic recession in Eastern Europe and accompanying drop in activity level had a factor 1.5 larger effect than policy measures taken in the western part on the overall sulphur reduction. Recent increases in fuel consumption in the recovering economies in Eastern Europe and also in Western Europe, is mainly from fuels which do not contain appreciable amount of sulphur. In addition, penetration of control technology all over Europe is reflected in a small but continuous decrease in European sulphur emissions.

p. 5126, line 24-27: For clarity I recommend to add average % numbers with the three periods, e.g. about 20% in the 80s, about 55% in the 90s and x% in the first half of the 2000s.

Response: Accepted. Three distinct emission regimes have been identified. During the first period from 1980 to 1989 emission reductions were generally low (20%), and largest in Western Europe. The highest emission reductions were reported in Eastern European countries during the second period, 1990-1999, characterised by high emission reductions (54%). The unification of Europe has lead to a more equally spread reduction pattern, with low-medium reductions all over Europe, and a total reduction of 17 % in the first half of the 2000s.

p. 5127, line 29-30: Phrasing is incorrect: .. SO₂ emissions it will provide a regional warming effect in Europe.

Response: We write in the manuscript p. 5103, line 19-21: The direct and indirect aerosol effects due to sulphate lead to a negative radiative forcing and thus a cooling effect on climate. Further we have emphasised on p. 5127, line 27-28 that: Local responses to a radiative effect are yet uncertain (Hansen et al., 2005). Hence we cannot fully accept the proposal for changes made by the referee, but will change the sentence on p. 5127, line 28-30 in the final version of the manuscript to: However, with such a strong reduction in the SO₂ emissions is it likely that this has contributed to a warming over Europe.

p. 5128, line 8: .. uses more detailed technology dependant emission factors and includes changes in technology mixes.

Response: We will correct the sentence in the final version of the manuscript to read:
p. 5128, line 8: This is probably due to the fact that the EMEP inventory uses more detailed country specific emission factors and includes changes in technology mixes.

p. 5128, line 9: .. importance of changes in technology mixes to estimate emissions ..

Response: We will correct the sentence in the final version of the manuscript to read:
p. 5128, line 8: From 1990 onwards the importance of capturing the changes in technologies by applying emission factors reflecting national circumstances becomes more evident in the comparison.

p. 5128, line 26: I miss here a conclusion on the importance of the non-regulated international shipping sources (e.g. present share in regional emissions and that their share increased and increases strongly). I also miss here a remark on the more strict NEC targets for EU countries.

Response: Please see response above on the NEC targets. With respect to the international shipping emissions we will add the paragraph below in the final version of the manuscript.

p. 5128, line 26: Emissions from international shipping, is not considered in this paper but clearly also influence the air quality and climate (e.g. Derwent et al., 2005; Marmer and Langemann, 2005). The trends in the ships emissions contrast the land based trends by an estimated increase of about 2.5% annually on cargo and 3.9% on passenger vessels in European waters (Cofala et al, 2007; Vestreng et al., 2006). Moreover, the SO₂ emissions from shipping are projected to increase by 42% from 2000 to 2020 in the current legislation baseline scenario (Cofala et al, 2007). Depending on ambition level, the emission from ships might stabilize or even decrease by as much as 70% in the case where maximum technological feasible reductions are con-

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sidered. Regional differences in trends are expected, as the sulphur emissions in the Baltic Sea and the North Sea are now regulated by the MARPOL Convention Annex VI (MARPOL, 1978) which entered into force in 2005.

Add to references: Cofala, J., Amann, M., Heyes, C., Wagner, F., Klimont, Z., Posch, M., Schöpp, W., Tarassón, L., Jonson, J. E., Whall, C., Stavrakaki, A., 2007. Analysis of Policy Measures to reduce Ships Emissions in the Context of the Revision of the National Emissions Ceilings Directive. Draft Final Report. European Commission, DG Environment, Unit ENV/C1. Contract No 070501/2005/419589/MAR/C1. Available from: <http://www.iiasa.ac.at/>

Derwent, R. G., D. S. Stevenson, R.M. Doherty, W.J. Collins, M. G. Sanderson, C. Johnson, J. Cofala, R. Mechler, M. Amann. and F. Dentener, 2005. The contribution from shipping emissions to air quality and acid deposition in Europe. *Ambio*, Vol. 34, No 1, February 2005.

Marmer, E. and B. Langmann, 2005. Impact of ship emissions on the Mediterranean summertime pollution and climate: A regional model study. *Atmos. Environ.* 39 (2005) 4659-4669, doi: 10-1016/j.atmosenv.2005.04.014

MARPOL (1978), International Convention for the Prevention from Ships, 1973, as modified by the Protocol of 1978 relating thereto. <http://www.imo.org/HOME.html>

p. 5136: Table 1. It is unclear to me why for a particular country the same gap filling/replacement applies to all years. Doesnt the actual coverage of sources per country vary over time?

Response: Whenever there is identified a need for gap filling or replacements, we seek to use the same independent estimates for the whole timeseries. It is impossible to mix e.g. EDGAR and RAINS data which is highly inconsistent as is EDGAR and reported data, and to a lesser extent RAINS and reported data. The independent estimates typically do not cover the whole period of investigation, hence interpolation

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and extrapolation is required, as stated also in the Table 1 heading.

Technical comments:

p. 5105, line 4: .. are national

p. 5111, line 7: I think deriving should be using

p. 5112, line 17: concentration measurements...

p. 5117, line 20: ..(FGD), mainly in power generation, ..

p. 5117, line 21: to gas or to nuclear power.

p. 5121, line 23: greenhouse gases

p. 5122, line 17: .. no substantial differences ..

p. 5128, line 7: .. in the other inventories.

Response: The above technical comments are appreciated and accepted. The referees suggestions will be incorporated in the final form of the manuscript

In addition to the above edits, we have ourselves found one typo which will be corrected in the final version of the manuscript. p. 5113, line 8-10 should read: Berglen et al. (2006) modelled 1985, 1995 and 2000 using the EMEP and Smith et al. (2004) inventories, together with the combined GEIA (<http://geiacenter.org>) / EDGAR (<http://www.mnp.nl/edgar/>) AeroCom (Dentener et al., 2006) dataset.

Add to References: Dentener, F., Kinne, S., Bond, T., Boucher, O., Cofala, J., Geroso, S., Ginoux, P., Gong, S., Hoelzemann, J., Ito, A., Marelli, L., Penner, J.E., Putaud, J.-P., Textor, C., Schulz, M., van der Werf, G.R., and Wilson, J.: Emissions of primary aerosol and precursor gases in the years 2000 and 1750 prescribed datasets for AeroCom, *Atmos. Chem. Phys.*, 6, 4321-4344, 2006.

Interactive comment on *Atmos. Chem. Phys. Discuss.*, 7, 5099, 2007.

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