

# Attributing a fraction of climate change to a nation's historical emissions: closure and scientific uncertainty

Version 24 October 2005

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**ACKNOWLEDGEMENTS****31****REFERENCES****31****1 Introduction**

(Mostly Copied from Paper # 1)

Observations of surface air temperature indicate that a significant global average warming has occurred during the 20th century. Several recent studies have attempted to partition this warming between natural and man-made causes (Tett et al., 1999; Mitchell et al., 2001) leading the Intergovernmental Panel on Climate Change (IPCC) to conclude that there is strong evidence that man has influenced the climate (IPCC, 2001). More recent studies (Stott, 2003; Zwiers and Zhang, 2003) have further strengthened this conclusion. Furthermore, climate models suggest that future man-made increases in greenhouse gas (GHG) concentrations will cause further climate changes (Cubasch et al., 2001; Johns et al., 2003), which could have large impacts on human and ecological systems (e.g. Parry, 2004). International negotiations have lead to a first step in combating climate change with the United Nations Framework Convention on Climate Change (UNFCCC) and the Kyoto Protocol, but further steps (i.e. emission reductions) are needed in order to achieve the ultimate objective of the UNFCCC of stabilizing the GHG concentrations at non-dangerous levels (UNFCCC, 1992).

The issue of allocating these reduction obligations amongst the regions, or Parties of UNFCCC, is called differentiation of future commitments. As part of the negotiations of the Kyoto Protocol, the delegation of Brazil presented one approach for allocating these reductions among OECD countries and economies in transition (the so-called Annex I Parties) based on the effect of their cumulative historical emissions of GHGs included in the Kyoto Protocol, from 1840 onwards, on the global average surface temperature (UNFCCC, 1997).

While the Brazilian Proposal was initially developed to further discussions on differentiation of commitments among Annex I countries, it can also be used as a framework for allocating emission reduction burdens across Annex I and non-Annex I countries. The proposal's central idea was that there exists a functional link between GHG emissions and global temperature increase, or other indicators along the cause-effect chain of climate change, such that the indicator can be calculated from the emissions using a simple model or set of models. The indicator acts as a surrogate for climate impacts, which are more difficult to calculate directly. The methodology also assumes that it is possible to apportion the contributions to the change in the indicator between a number of sources and emitters (e.g. nations, regions).

Although it was not adopted during the Kyoto negotiations, the Brazilian Proposal did receive support, especially from developing countries, and the Third Conference of the Parties (COP-3) requested the Subsidiary Body on Scientific and Technical Advice (SBSTA) to further study the methodological and scientific aspects of the proposal. This led to continued debate and analysis (e.g. Enting, 1998; Filho and Miguez, 1998; den Elzen et al., 1999; den Elzen and Schaeffer, 2002; Höhne, 2002; Rosa et al., 2004) and a number of expert meetings organized by the UNFCCC secretariat. The objective of these meetings was to review the scientific and methodological aspects of the proposal and to co-ordinate an intercomparison of attribution results using a set of simple climate models in an exercise called the Assessment of Contributions to Climate Change (ACCC). The conclusions of this analysis are described in

UNFCCC (2002b), and some institutes have reported their analysis in more detail (e.g. den Elzen et al., 2002; Andronova and Schlesinger, 2004; den Elzen et al., 2005; Höhne and Blok, 2005; Trudinger and Enting, 2005).

A follow up exercise is now being carried out by an ad hoc group for the modeling and assessment of contributions to climate change (MATCH) (Höhne and Ullrich, 2003) to improve the robustness of calculations and more rigorously assess the uncertainties and methodological choices. In the first joint paper (den Elzen et al. 2005), the results from the first activities of MATCH are summarized, and the central questions are addressed: how robustly can simple climate models (SCMs) be used to attribute anthropogenic climate change to sources of GHGs (e.g. regional sources) and what effect do a range of scientific choices (related to scientific uncertainties) and policy-related choices, that are part of the negotiation process, have on these attribution calculations?

An approach based solely on rationing the reported accumulated emissions of the Kyoto gases from Annex I countries has some potential, hidden biases. Simply put, there is a closure problem: our best models do not always match the observed climate system changes, including the greenhouse gas abundances; and thus uncertainties implied in this mismatch of absolute changes must be transferred to uncertainty of relative changes. Without some level of confidence that (i) all known emissions of greenhouse gases (anthropogenic and natural) can explain the historical record of atmospheric abundance and (ii) this observed history of greenhouse gases plus other radiative forcings (e.g., solar, volcanoes, and especially anthropogenic aerosols) can explain the observed climate changes over the 20<sup>th</sup> century, there are possible errors in the national emissions that may be country specific, even within Annex I. This second joint paper of the MATCH group examines the level of closure in items (i) and (ii) above insofar as it points out the magnitude of unspecified sources or sinks for greenhouse gases and aerosols. It compares these with uncertainties derived from bottom-up national inventories such as EDGAR-HYDE. We then follow the propagation of both measures of scientific uncertainty as in the first paper (den Elzen et al. 2005) to two simple indices of climate change, the global mean surface temperature (T<sub>surf</sub>) and the sea-level rise due to oceanic heat uptake (O<sub>cn</sub>).

Paper #2 combines the propagation of errors in accountable emissions with closure calculations to estimate possible biases, such as over/under-counting greenhouse gas emissions, or other radiative forcings, that might be country specific. It examines the scientific uncertainty associated with attributable emissions.

[Reference to other similar papers]

[Outline of the paper]

## **2 Long-lived greenhouse gases**

In this chapter, we analyse the uncertainties in Annex-I emissions from inventories for the long-lived greenhouse gases as well as possible global bias in reconciling emissions and measured abundances of these gases in the atmosphere.

For these long-lived components of Radiative Forcing (RF) we can use the observed atmospheric record (whole air, firn air, and Ice cores) to define the RF history. Thus, our questions here are separated into the "attribution" one (what are Annex-I emissions?) and the overall "closure" one (is there a systematic bias between emissions, models, and observed abundances?).

## 2.1 CO<sub>2</sub>

### 2.1.1 Annex I emissions by country

(From A. Kurosawa, August 25, 2005)

The UNFCCC Secretariat has summarized Annex-I countries annual GHG inventory reporting data since 1990. The most recent database (UNFCCC, 2005) covers emissions from these countries from 1990 to 2002. IEA (2004) and CDIAC (Marland, et.al., 2003) also reported country-based emission data of most world countries. There are two IEA kinds of assessment data based on reference and sectoral method. The former uses energy supply information while the latter depends on demand information.

IPCC (2000) has discussed uncertainty in terms of activity level, emission factor and estimation method and concluded overall uncertainty of CO<sub>2</sub> emission from fossil fuel combustion uncertainty is likely to be less than 5% for traded fuels and higher than for non-traded fuels. IEA (2004) has made comparison of UNFCCC National Communication and own estimate from methodological point of view. It showed that for most Annex I countries two calculations were within 5%, while some EIT and Non-Annex I countries' difference tended to be larger.

We compiled and compared data from IEA reference, IEA sectoral and CDAIC with UNFCCC inventory data by country after 1990 to check the consistency among them. Numbers of IEA reference approach are always larger than those of sectoral approach since methodological difference lies in the estimation process. UNFCCC emissions data as well as maximum and minimum differences between UNFCCC and other data in 1990 and 2000 are summarized in Figure 1 and Figure 2.

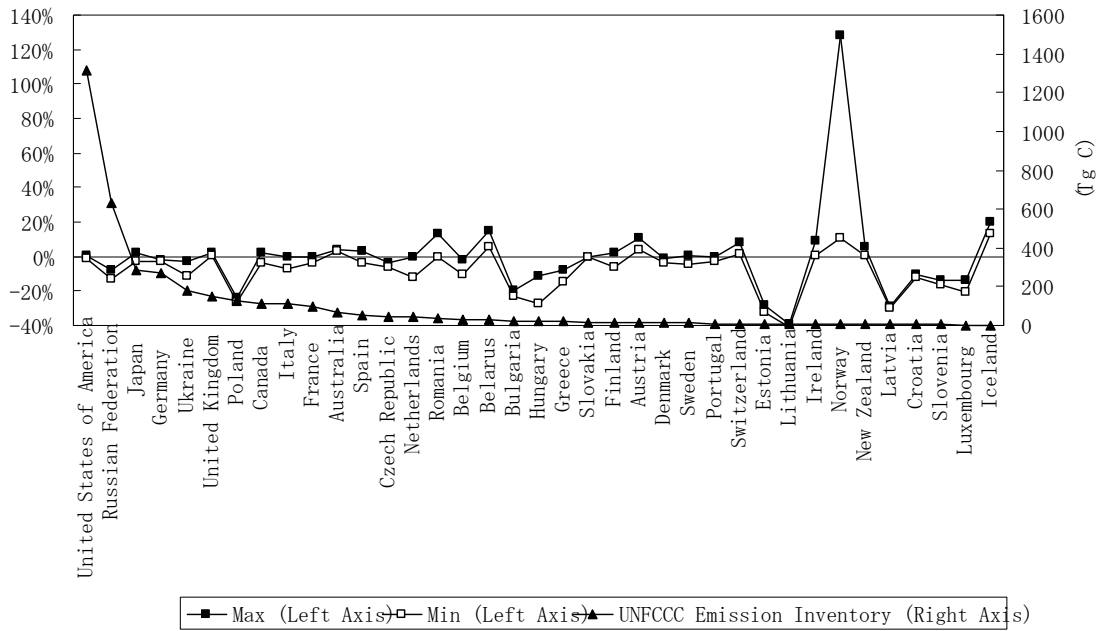


Figure 1 UNFCCC fossil fuel combustion CO2 emissions inventory and database uncertainty range of Annex I countries in 1990

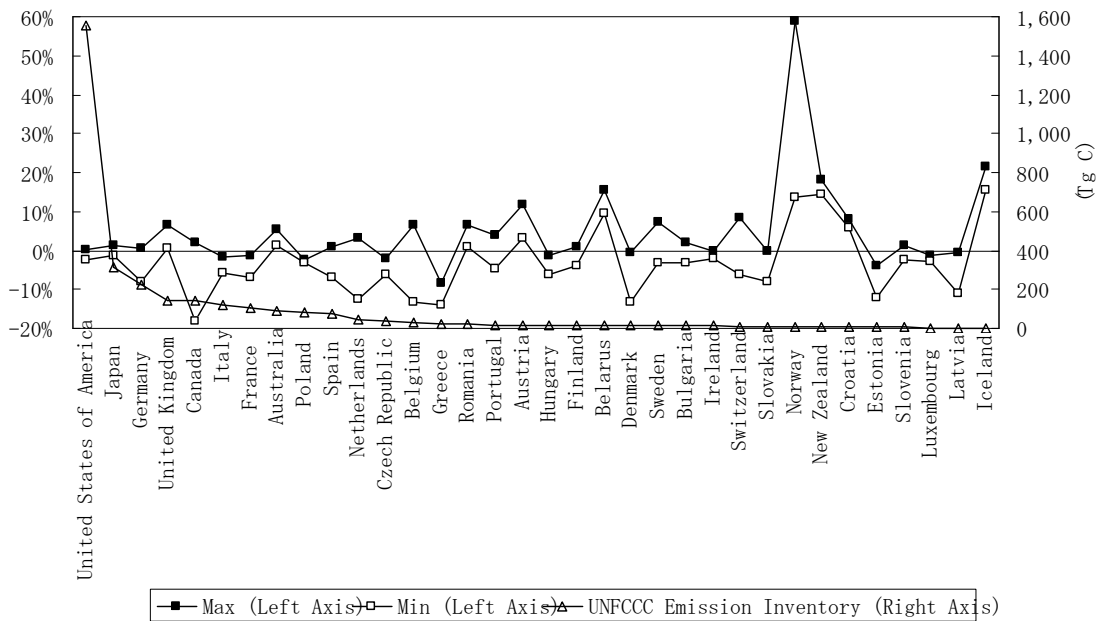
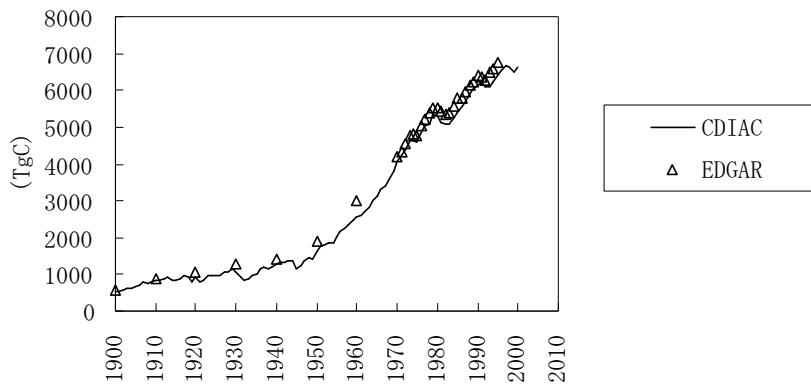


Figure 2 UNFCCC fossil fuel combustion CO2 emissions inventory and database uncertainty range of Annex I countries in 2000

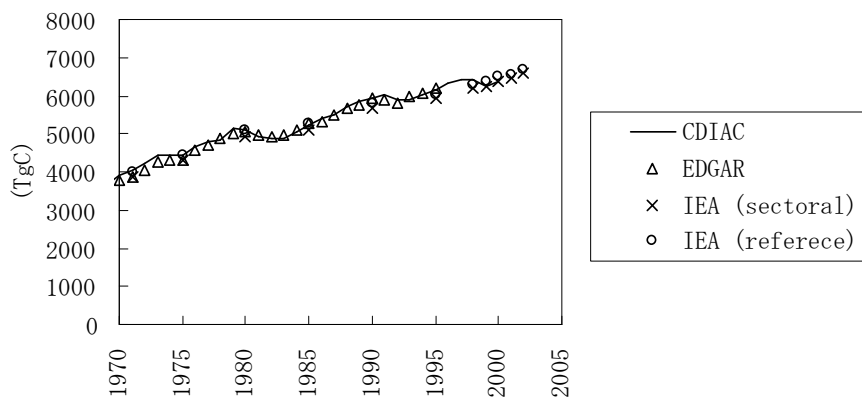
The result from existing literatures and the database intercomparison above leads to the conclusion that Annex-I country level database uncertainty range of recent years is less than 5 to 10% with minor exceptions.

We made another comparison of existing global fossil CO<sub>2</sub> emission databases. Total CO<sub>2</sub> emissions from fossil fuel including fugitive emission, emission from combustion, and industrial process are shown in Figure 3 extracted from EDGAR (Oliver, 1999) and CDIAC. The uncertainty range of two databases is approximately 10% before 1970. On the contrary, the range decreases to approximately 5% after 1970 reflecting the high consistency level of fossil fuel combustion emission.



**Figure 3 Global CO<sub>2</sub> emissions from fossil fuel**

Figure 4 indicates fossil fuel combustion emission numbers from EDGAR, CDIAC and IEA after 1970. The maximum range of uncertainty from four databases is approximately 5%.



**Figure 4 Global CO<sub>2</sub> emissions from fossil fuel combustion**

**2.1.2 Possible biases / lack of understanding of global CO<sub>2</sub> emissions**  
(Jain, de Campos, Kant)

### 2.1.3 Discussion

(A. Kurosawa, August 25, 2005)

We have to pay attention to the cause of emissions data uncertainty. IPCC guideline (1997) provided default method (Tier 1) and bottom-up method (Tier 2 and 3) for preparing national emission inventory report for Annex I countries. It recommended that countries which have used a detailed sectoral approach for CO<sub>2</sub> emissions from energy combustion also use the reference approach for verification purposes. IEA (2004) compared two approaches and pointed out the possibility of significant discrepancies between them. It stated that the origins of discrepancies are statistical differences between supply and demand, aggregate net calorific values and emission factors, allocation of fuel inputs to the transformation sector excluding power and heat generation, misallocation of fuels to the energy sector, missing information on certain transformation outputs, distributions losses, and transfers and reclassification of energy products. It also pointed out that the reasons of differences lies in the treatment of international bunker, energy activity data, net calorific values, emission factors, stored carbon, military emissions, coke categorization, temperature adjustment and electricity trade. It illustrated the examples of country level discrepancies between sectoral and reference approach with possible causes in 1990 and 2002 data for Czech Republic (statistical difference for coking coal, carbon inflows to gas works plants), Hungary (statistical difference for lignite and gas/diesel oil, carbon flows to/from transformation process), Japan (coke ovens and statistical difference), New Zealand (fugitive losses for liquefaction plants and statistical differences), Norway (statistical difference of natural gas), Russia (distribution losses, blast furnaces), Sweden (oil refineries and coke ovens efficiencies) and Turkey (coke ovens and oil refineries). Finland, France, Iceland, Ireland and Switzerland were countries which included in the report for their difference by statistical differences.

[Discussion possible biases]

## 2.2 CH<sub>4</sub>

### 2.2.1 Annex I emissions by country

(Version 06.04.05 , Fabian Wagner)

Methane emission data are typically more uncertain than CO<sub>2</sub> emission data from fossil fuels, primarily due to large uncertainties in the emission factors. The emission factors are more uncertain first because for methane, unlike for CO<sub>2</sub>, national emission factors are often unavailable and highly aggregated regional default factors are applied. Moreover, methane emission factors are often highly dependable on actual circumstances of the source. For example, emission rates from livestock will depend on dietary composition, which may not be uniform across a country or region.

Activity data are typically associated with less uncertainty than the emission factors as they are often derived from commodity statistics (e.g. livestock, coal and gas), but may also be considerably uncertain (e.g. carbon stocks in landfills, amount of degradable organic carbon deposited, etc.).

The overall uncertainty in emission estimates at the national or regional scale depends on the uncertainty of individual source categories and their relative contribution to the national total. According to UNFCCC (2003) in 2001 more than 35 percent of all reported methane emissions in Annex I Parties (57 Mt CH<sub>4</sub>/year) stem from livestock (mostly enteric fermentation from ruminant animals) ranging from 18 percent (Bulgaria) to 88 percent (New Zealand) of the

national total. The second largest source category across all reporting parties was solid waste disposal sites (33 percent of all reported Annex I methane emissions), and the third largest is fugitive emissions in the energy sector (27 percent; including pipeline transport and coal bed methane). These contributions to the total national methane emissions, however, also vary considerably across parties (for landfills: from 6 percent in Monaco to 57 percent in Finland; for fugitive: from 0 percent in Sweden to 54 percent in the Czech Republic).

For many Asian countries and some others rice cultivation is also a major source of methane emissions; e.g. in Japan about 30 percent of the total methane emissions origin in rice paddies. Most other sources are reported to be only a small part of the national total (methane emissions from combustion are reported at approximately 10 percent of the national total in some European countries).

IPCC (2000) has provided default uncertainty values for some methane emission factors and model parameters. The uncertainty default values largely depend on whether emissions are estimated using the default (Tier 1) method or national emission factors/national methods (Tier 2/3). The IPCC estimates for the emission factor uncertainties are given in Table 1:

**Table 1 IPCC default uncertainties for methane emission factors for the main source categories. (Source: IPCC (2000))**

<b>Source</b>	<b>Tier 1</b>	<b>Tier 2/3</b>
Livestock	30-50%	20%
Coal Mine - Underground	50-75%	Factor of 2
Coal Mine - Surface	Factor of 2	Factor of 3
Coal Mine – Post Mining	50%	Factor of 3
Oil and Gas Operations	Order of magnitude	25-50%
Stationary Combustion	50%	N.A.
Landfill	N.A. <sup>(1)</sup>	N.A.

- (1) Depends on uncertainty in fraction of degradable organic carbon and its fraction dissimilated, the uncertainty in the methane correction factor, methane recovery and oxidation factor. Finally, the generation rate constant has an uncertainty of approximately a factor of 3.

Uncertainties in the models that are used in the estimation of greenhouse gas emissions are not covered in IPCC (2000).

### Estimated vs Reported Emissions - CH<sub>4</sub>

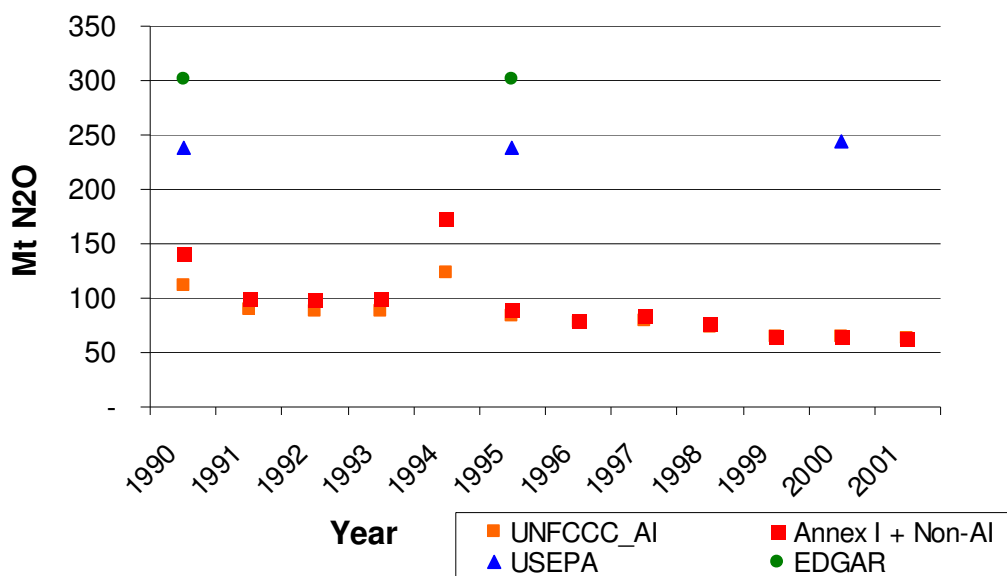


Figure 5 Estimated versus reported Emissions – CH<sub>4</sub>

Most Annex I countries have submitted to the UNFCCC not only emission estimates but also uncertainty estimates by source category and gas. For this study we have analyzed the uncertainty estimates in total methane emissions of selected Annex I countries (Finland, Germany, Japan, UK). These range from 15-30 percent of the national total. The highest uncertainty in methane emissions is typically associated with the estimates of emissions from landfills.

The documentation of the EDGAR 3 database (*Olivier et al., 1999*) provides qualitative indications of uncertainties in the activity data, emission factors and total emissions from various sources. For methane *Olivier et al. (1999)* conclude that their national total emission estimates is associated with medium uncertainty, viz. approximately 50 percent. That this value lies higher than the typically reported values by Annex I parties is due to the fact that for EDGAR mostly IPCC Tier 1 methods were used to estimate emissions, while Annex I parties oftentimes employ higher tier methods. It can be expected that also for non-Annex I parties overall uncertainties amount to more than 30 percent since for lack of resources typically Tier 1 methods are used.

Uncertainties in the national estimates cannot be combined into an uncertainty in the global emission estimate using a Tier 1 method for combining uncertainties as it is used at the national level. This method does not take into account that countries may be (and should be) using similar methods and parameter values, so that emission estimates are correlated. This may lead to a bias at the global scale. As an alternative, a weighted average of uncertainties of the larger emitter countries can be used as a default for the uncertainties in the global emission estimate.

#### 2.2.2 Possible biases / lack of understanding of global CH<sub>4</sub> emissions

(Fuglestad, Prather)

Using a simple box model of the global atmosphere we have calculated the development in concentration of methane,  $C(t)$ , for various assumptions about lifetime/adjustment time. These calculated concentrations are then compared to *observed* concentrations. What this means in terms of deviations in emissions are finally calculated. We have used emissions numbers used in MATCH paper #1 (based on EDGAR) and assumed constant natural emissions equal to 293 TgCH<sub>4</sub>/yr.

The main loss mechanism for methane is reaction with the OH radical in the troposphere. Loss to the stratosphere and uptake in soil and loss to the stratosphere also contribute to the total loss, which can be given as  $1/\tau_{\text{tot}} = 1/\tau_{\text{OH}} + 1/\tau_{\text{strat}} + 1/\tau_{\text{soil}}$

The “best estimate” in IPCC TAR is given as:  $1/8.4 = 1/9.6 + 1/120 + 1/160$

We have calculated development in methane concentrations for various assumptions about the tropospheric loss rates based on reported values in the literature (i.e. the soil and stratospheric losses have been kept constant, or left out). The following cases are considered, where the first seven cases use constant loss of methane over time, while the rest allow variations in the tropospheric loss rate:

**Table 2. Various assumptions about loss of methane used in the calculations.**

Case	ASSUMPTION	$\tau_{\text{tot}}$ (years)	Comment	Reference
1	$1/\tau_{\text{tot}} = 1/13.8 + 1/120 + 1/160$	11.5	Max value for $\tau_{\text{OH}}$ from table 4.3 in TAR	TAR, table 4.3
2	$1/\tau_{\text{tot}} = 1/12 + 1/120 + 1/160$	10.2	12 years is adjustment time including CH <sub>4</sub> -OH feedback	TAR, table 6.7
3	$1/\tau_{\text{tot}} = 1/9.6$	9.6	No soil or stratospheric loss assumed.	
4	$1/\tau_{\text{tot}} = 1/9.6 + 1/120 + 1/160$	8.4	IPCC “default case”	TAR
5	$1/\tau_{\text{tot}} = 1/6.5 + 1/120 + 1/160$	5.9	Min value for $\tau_{\text{OH}}$ from table 4.3 in TAR	TAR, table 4.3
6	$1/\tau_{\text{tot}} = 1/9.6 \cdot 1.4 + 1/120 + 1/160$	11.2	The tropospheric loss is increased by 1.4 due to CH <sub>4</sub> -OH feedback	TAR
7	$1/\tau_{\text{tot}} = 1/10.12 + 1/120 + 1/160$	8.8	10.12 years from:	Prinn et al., 2001
8	$\tau_{\text{OH}}(t)$ calculated as function of CH <sub>4</sub> concentration and emissions of NO <sub>x</sub> , CO and VOC	Variable (see fig 1)		TAR, table 4.11, footnote b
9	$\tau_{\text{OH}}(t)$ calculated as function of CH <sub>4</sub> concentration	Variable (see fig 1)	The effect of NO <sub>x</sub> , CO and VOC on OH is ignored.	TAR, table 4.11, footnote b
10	$\tau_{\text{OH}}(t)$ calculated as function of CH <sub>4</sub> concentration and emissions of NO <sub>x</sub> , CO and VOC. Natural emissions: 150 Tg/yr	Variable		TAR, table 4.11, footnote b
11	$\tau_{\text{OH}}(t)$ calculated as function of CH <sub>4</sub> concentration and emissions of NO <sub>x</sub> , CO and VOC. Model start: 1890	Variable		TAR, table 4.11, footnote b

Figure 6 shows the development in total adjustment time  $1/\tau_{tot}$  (when the TAR formulation is used for  $\tau_{OH}(t)$ ; labelled tau-TAR) and without emissions of the ozone precursors NOx, CO and VOC (labelled tau-TAR no O3P):

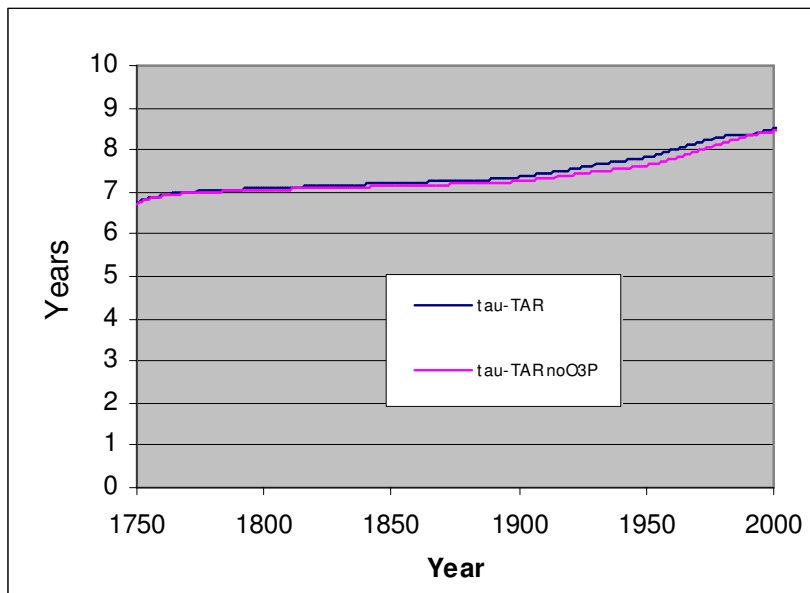


Figure 6. Development in  $\tau_{tot}$  for cases with and without NOx, CO and VOC.

Based on the assumptions given in table 1 the developments in methane concentrations are calculated and shown together with observed methane concentrations, see Figure 7.

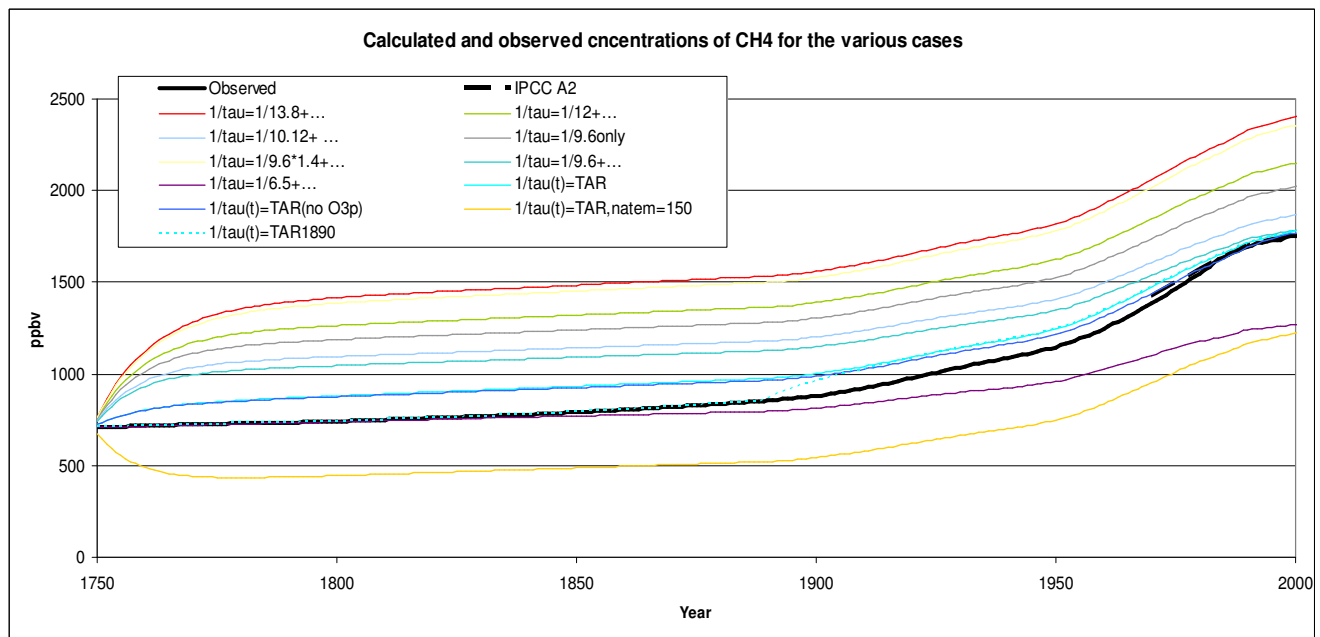
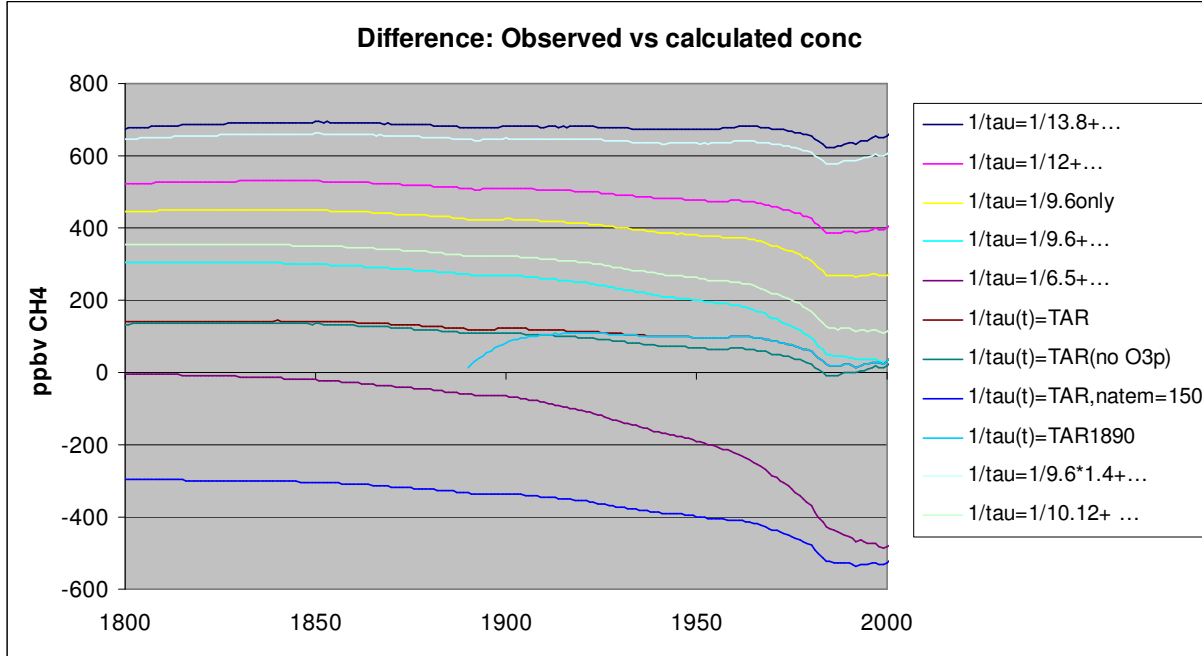


Figure 7. Calculated and observed concentrations of methane. Observed concentrations are taken from: [www.giss.nasa.gov/data/simodel/ghgases/Fig1C.ext.txt](http://www.giss.nasa.gov/data/simodel/ghgases/Fig1C.ext.txt)

Figure 8 shows the differences between observed and calculated concentrations.



**Figure 8. Differences between calculated and observed concentrations of methane.** (Possibly skipped)

The next step is to calculate the implied emission deviation. We use the general formula for the time development of a gas:

$$dC/dt = P - L \cdot C = P - C/\tau \quad (1)$$

Where

C: Conc

P: Production rate

L: Loss rate

$\tau = 1/L$ : lifetime

Rearranging (1) we obtain

$$P = dC/dt + C/\tau \quad (2)$$

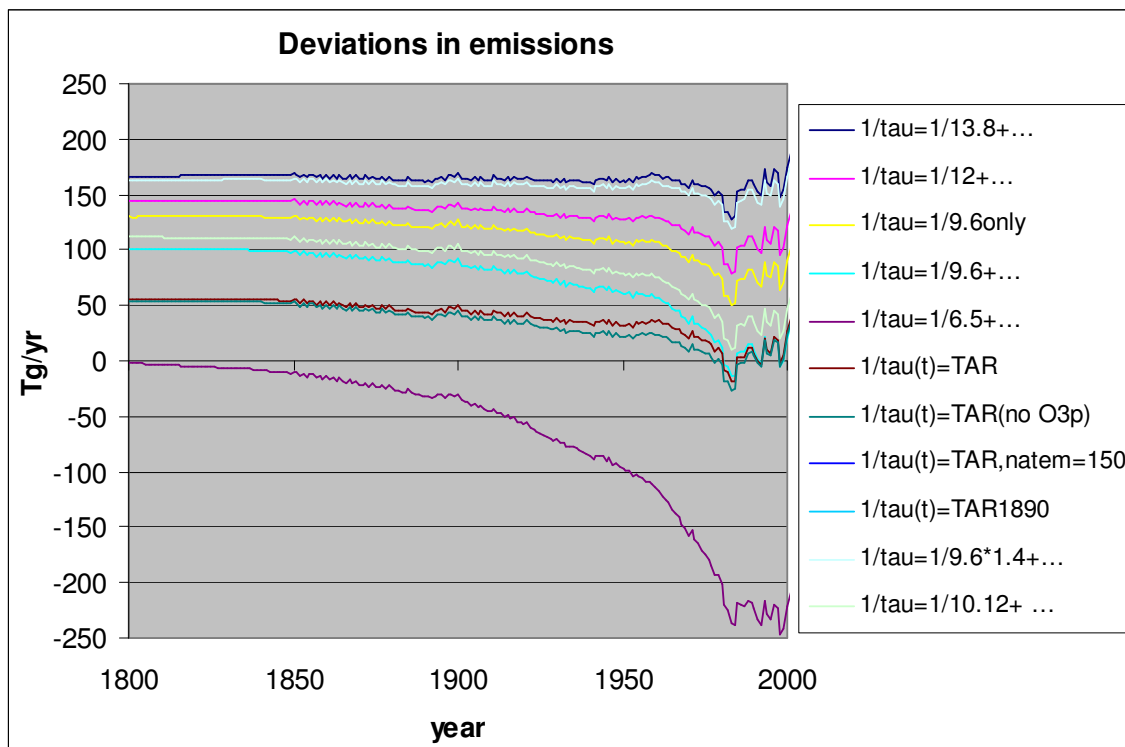
In order to calculate the deviation in production between calculated and observed concentration, we use:

$$\Delta P = d\Delta C/dt + \Delta C/\tau \quad (3)$$

where  $\Delta P$  is deviation in production,  $\Delta C$  is deviation in concentration and  $d\Delta C/dt$  is development of this deviation. To obtain annual emissions, we multiply with a conversion factor  $\beta$ :

$$\Delta E \text{ (Tg/yr)} = \Delta P(\text{ppbv/yr}) \cdot \beta \text{ (Tg/ppbv)} = \beta (d\Delta C/dt + \Delta C/\tau)$$

By this approach we calculate the following time dependent deviation in emissions for the various cases:

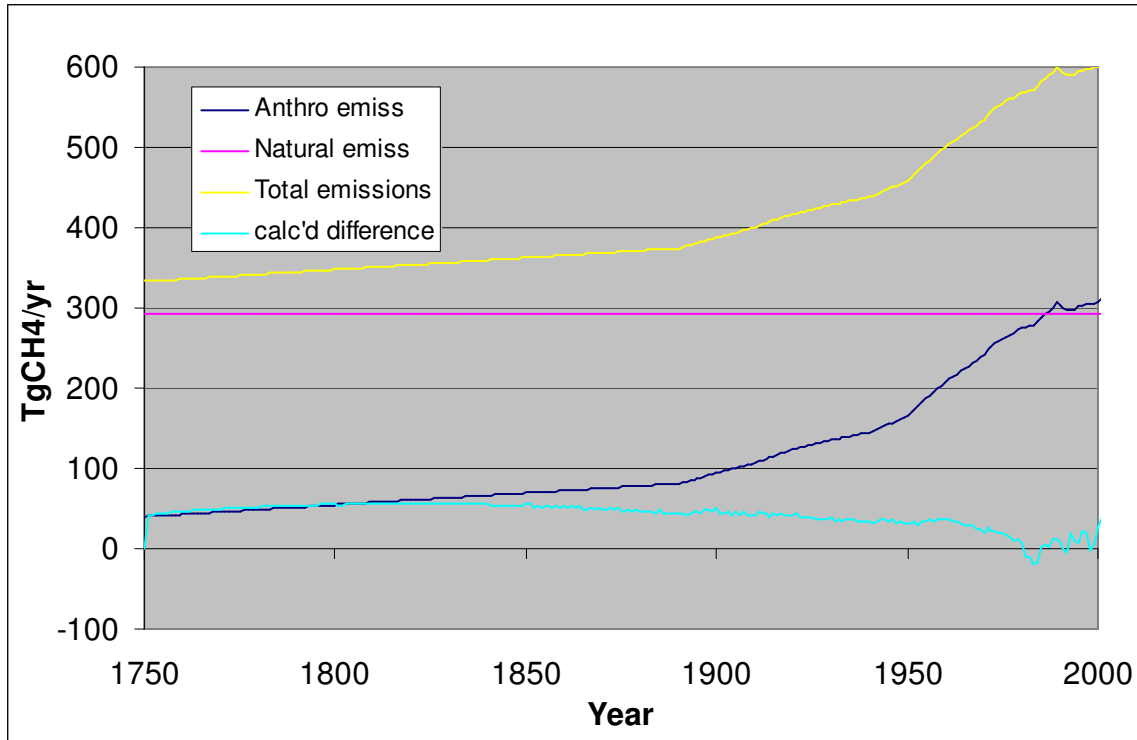


**Figure 9. Inferred deviations in emissions that can explain the deviations in concentrations between calculated and observed concentrations. (The case ‘TAR and natural emissions = 150’ is not included here due to lack of realism; see figure 3)**

These calculations show that: the cases using the TAR formulations give the best estimates for the development of the global atmospheric concentration.<sup>1</sup> Assuming lower natural emissions would improve the estimates.

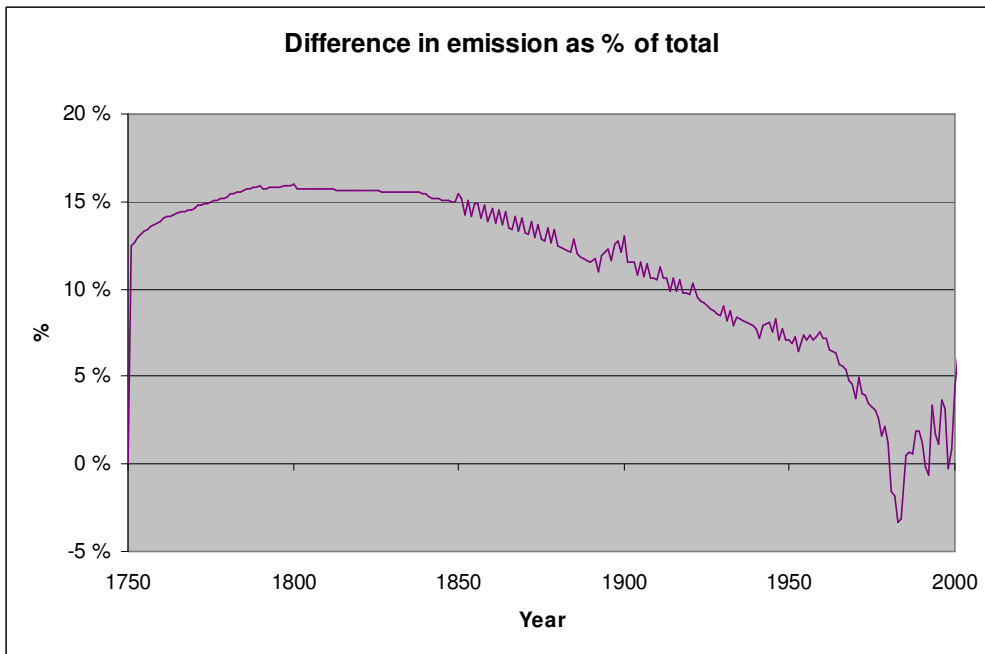
Figure 10 shows the development in anthropogenic, natural and total emissions together with the calculated deviation in emission for the case were the TAR formulation is used for OH loss (tau-TAR case).

<sup>1</sup> As a check we have adjusted the emissions by these inferred deviations (i.e. used these as a perturbation to the input emissions for the box model) and as expected, a very good agreement with observations is found, although not identical due to the CH<sub>4</sub>-OH feedback. Iterations would make the difference converge towards zero.



**Figure 10. Development in anthropogenic, natural and total emissions, and calculated deviation in emission.**

For this choice of  $\tau$  (t), the magnitude of the deviation in emissions compared to the total emission is less than 15% after 1850 and approaching just a few percent during the later part of the 20<sup>th</sup> century; see Figure 11.



**Figure 11. Difference in emissions as percentage of total emission for the tau-TAR case (possibly skipped)**

Compare this uncertainty with the uncertainty in emissions:

Based on input so far:

Uncertainty in anthropogenic emissions: 50%

Uncertainty in natural emissions: 50% (CHECK & DISCUSS THIS !)<sup>2</sup>

$$(\sigma E_{\text{tot}})^2 = (\sigma E_{\text{n}})^2 + (\sigma E_{\text{a}})^2$$

where

$E_{\text{tot}} = E_{\text{n}} + E_{\text{a}}$

$E_{\text{n}}$ : Natural emissions

$E_{\text{a}}$ : Anthropogenic emissions

$\sigma$ : standard deviation

If natural emissions uncertainty is 50%, total uncertainty range is 35% by the following.

$$\sqrt{(146.5 \cdot 146.5 + 150 \cdot 150)} / (293 + 300) = \sim 0.35$$

### 2.2.3 Discussion

[Use this for evaluation of the quality of the inventories and possible systematic error in emissions:

- Are there some missing sources?
- Underestimates / overestimates of source strengths

The history of the deviation may give important info about where the errors are.

Gives us qualitative assessment of the level of understanding of the gas budget. But applies only to **total global emissions** (natural + anthropogenic) and represents a fundamental scientific uncertainty in our understanding.

Discuss briefly whether this/these errors could be within Annex I, RoW or natural sources, if we can.

Are the errors confined to some specific sectors? This overlaps with section 2.1.

If so, this may affect the national emissions differently and thus the attribution calculations. Use expert judgements.]

- Many possible formulations lead to a misfit between resulting and observed concentrations
- Within the uncertainty no major sources missed
- Sum of national emission estimates are plausible given the observed concentrations and uncertainty. No systematic bias for the contribution calculations.
- A reasonable good agreement with observations is obtained with a simple box model using the TAR expression for lifetime as function of CH<sub>4</sub> concentration and emissions of NO<sub>x</sub>, CO and VOC.

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<sup>2</sup> The wetland CH<sub>4</sub> uncertainty in TAR is around +/- 100 TgCH<sub>4</sub>/yr and could be a good proxy for natural emission uncertainty.

## 2.3 N<sub>2</sub>O

### 2.3.1 Annex I emission by country

(Version 06.04.05 , Fabian Wagner, wagnerF@iiasa.ac.at)

Uncertainties in national N<sub>2</sub>O emission estimates are typically higher than those of CH<sub>4</sub> emissions, especially uncertainties in the emission factors. This is due to the fact that the underlying processes leading to N<sub>2</sub>O emissions are typically less well understood. In particular, estimating indirect emissions following leaching and runoff as well as volatilization of N-compounds requires significant knowledge of the local characteristics of N-cycle processes. Moreover, emission factors can be highly dependent on specific circumstances (e.g. combustion temperature, effectiveness of NO<sub>x</sub> control measures, etc) and confidence intervals are often skewed and long-tailed. As a consequence, applying an average emission factor may lead to substantial over- or underestimation of the actual emissions.

According to UNFCCC (2003) in 2001 almost 70 percent of all reported nitrous oxide emissions in Annex I Parties were generated in the agricultural sector (agricultural soils, manure management) ranging from 43 percent (Belgium, Bulgaria) to 96 percent (New Zealand) of the national total. Emissions from transport and industrial processes account for about 10 percent each of all N<sub>2</sub>O emissions that were reported by Annex I parties for 2001 (3027 kt N<sub>2</sub>O/year), while fugitive emissions and emissions from combustion in the energy sector accounted for approximately 7 percent. Again individual sectors' contribution to the national total vary widely across reporting Annex I parties.

In non-Annex I countries emissions from savanna burning may be a substantial source of N<sub>2</sub>O, now and in the past.

In the following table default uncertainties in N<sub>2</sub>O emission factors ((IPCC (2000), often based on expert judgment)

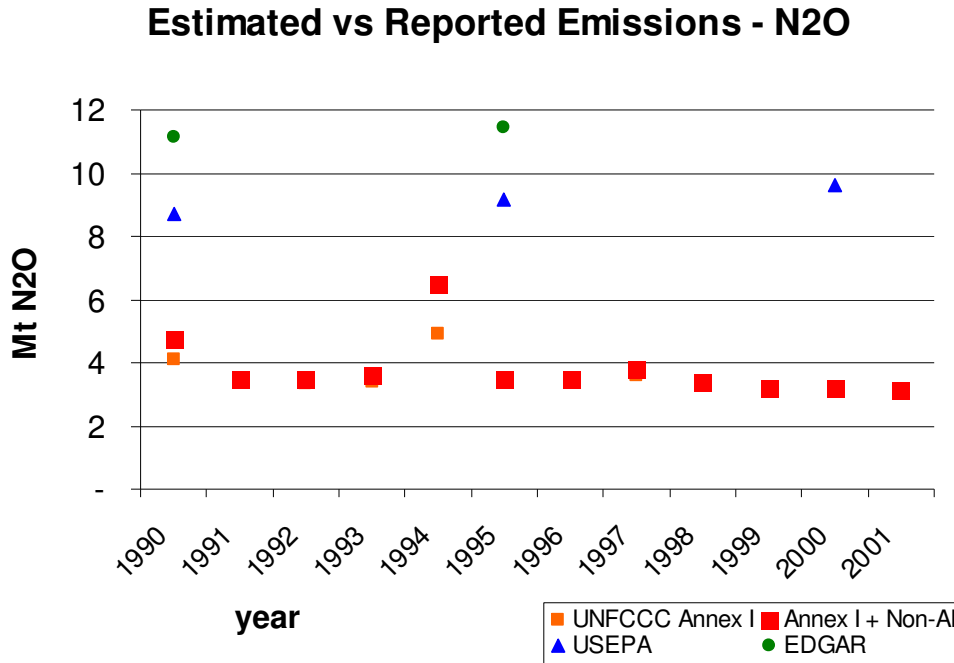
**Table 3 IPCC default uncertainties for nitrous oxide emission factors for the main source categories. (Source: IPCC (2000))**

Source	Tier 1 - Uncertainties
Stationary Combustion	Order of magnitude
Mobile Combustion	> 50%
Adipic and nitric acid production	> 5%
Manure Management	Factor of 2
Direct soil emissions	Factor of 5
Indirect soil emissions	Order of magnitude

On the basis of National Inventory Reports for the 2004 submission to the UNFCCC we have estimated the uncertainties in the national total emission of N<sub>2</sub>O for selected Annex I parties. While these uncertainties are estimated for several parties to lie between 30 and 40 percent, the estimated value for the overall uncertainty reported by the UK lies more than an order of magnitude higher. This is due to the fact that the uncertainty values assumed by the UK for

individual sources of N<sub>2</sub>O are at the upper end of the values in the table above. In particular, soil emissions are assumed to have indeed an order of magnitude uncertainty and are significant. Whether this is an adequate description of the actual uncertainty is disputable.

*Olivier et al(1999)* conclude that the EDGAR 3 total emission estimates are associated with large uncertainty, viz. 100 percent (factor of 2).



**Figure 12 estimated versus reported Emissions – N<sub>2</sub>O**

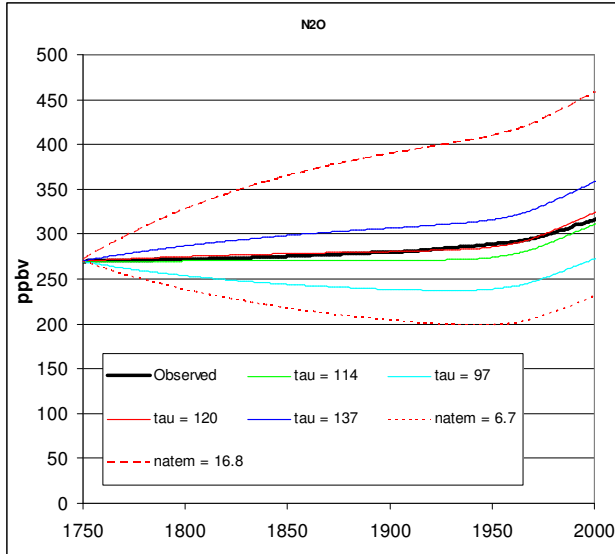
The caveat mentioned above on combining uncertainties in the national emission estimates to an uncertainty of the global emissions apply for N<sub>2</sub>O as well.

### 2.3.2 Possible biases / lack of understanding of global N<sub>2</sub>O emissions

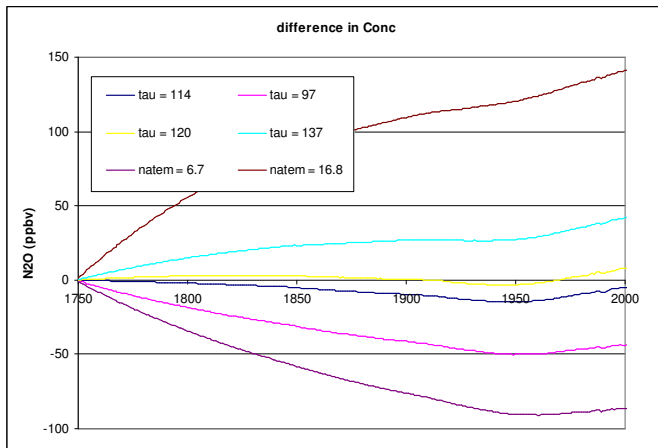
(Jan Fuglestedt, Bård Romstad, Michael Prather)

Using a simple box model of the global atmosphere we have calculated the development in concentration of N<sub>2</sub>O, for various assumptions about lifetime and natural emissions. These calculated concentrations are then compared to *observed* concentrations. What this means in terms of deviations in emissions are finally calculated. We have used emissions numbers used in MATCH paper #1 (based on EDGAR) and assumed constant natural emissions equal to 10.7 TgN/yr.

For various assumptions on lifetime and natural sources (see legend) the developments in concentrations are calculated and shown together with observed concentrations, see Figure 13. Figure 14 shows the differences between observed and calculated concentrations.

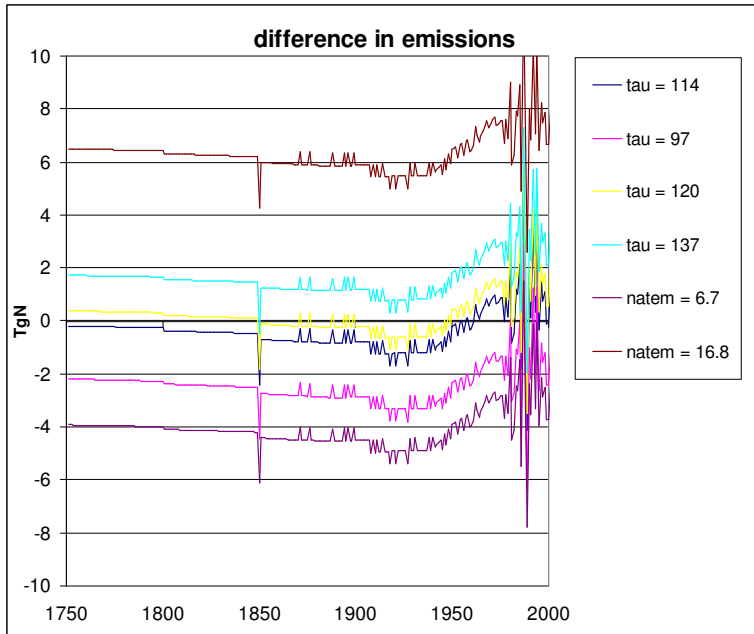


**Figure 13** Calculated and observed concentrations of N<sub>2</sub>O. Observed concentrations are taken from: [www.giss.nasa.gov/data/simodel/ghgases/Fig1C.ext.txt](http://www.giss.nasa.gov/data/simodel/ghgases/Fig1C.ext.txt)



**Figure 14.** Differences between calculated and observed concentrations of N<sub>2</sub>O. (maybe we will skip this one)

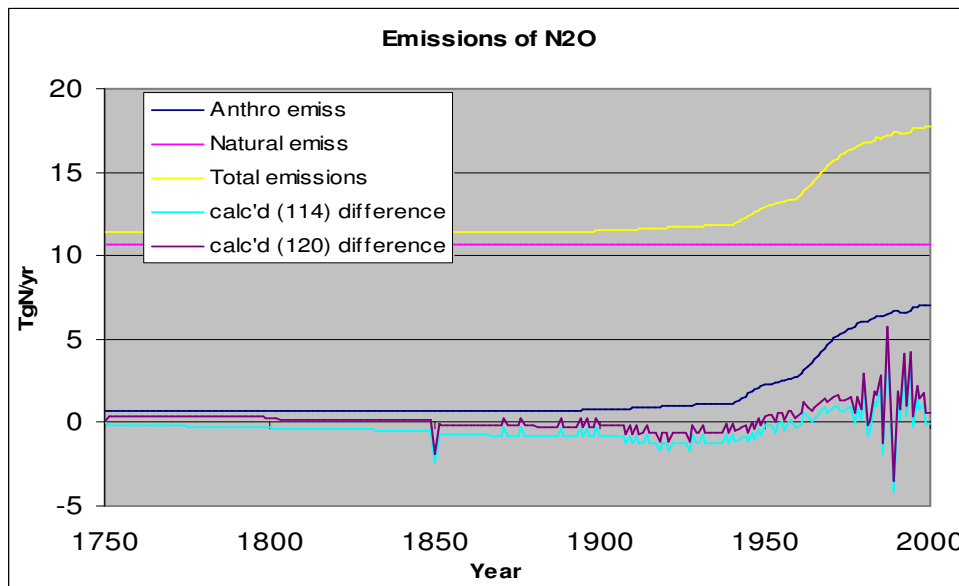
We have used the same approach as in section 2.2.2 to calculate the implied emission deviation for the various cases, see Figure 15.



**Figure 15. Inferred deviations in emissions that can explain the deviations in concentrations between calculated and observed concentrations.**

These calculations show that the cases using 114 and 120 years as lifetime give the best estimates for the development of the global atmospheric concentration.

Figure 16 shows the development in anthropogenic, natural and total emissions together with the calculated deviation in emission for the case were 114 or 120 years is assumed for the lifetime of N<sub>2</sub>O.



**Figure 16. Development in anthropogenic, natural and total emissions, and calculated deviation in emission**

For these cases, the magnitude of the deviation in emissions compared to the total emission is less than 10% before 1970 and increasing to around 20% during the later part of the 20<sup>th</sup> century; see Figure 17.

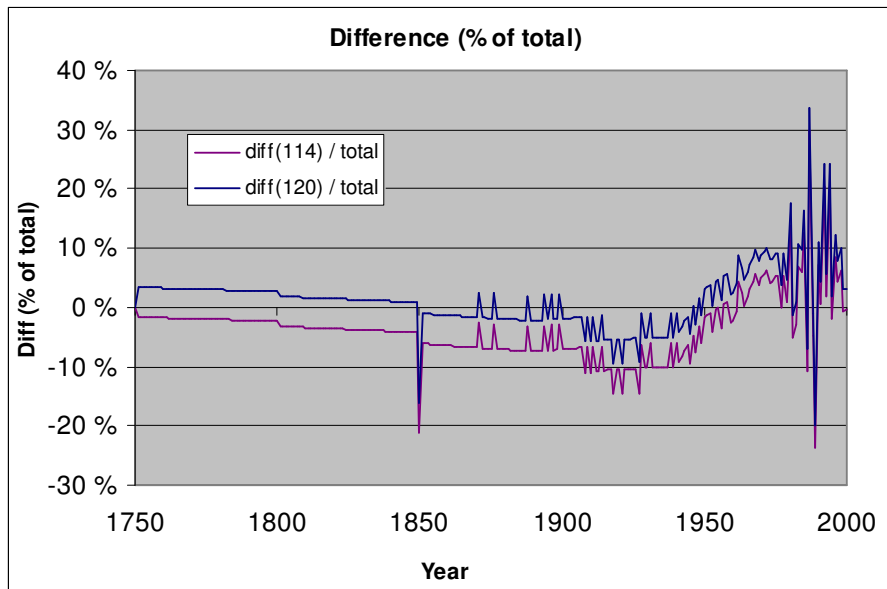


Figure 17. Difference in emissions as percentage of total emission for two cases.

### 2.3.3 Discussion

Based on input so far:

Uncertainty in anthropogenic emissions: 7 TgN/yr; i.e. 100%

Uncertainty in natural emissions: 5 TgN/yr; i.e. 50% (Needs to be discussed)

Implies 50% uncertainty in total emission.

[Use this for evaluation of the quality of the inventories and possible systematic error in emissions:

- missing sources?
- Underestimates / overestimates of source strengths

The history of the deviation may give important info about where the errors are.

Gives us qualitative assessment of the level of understanding of the gas budget. But applies only to **total global emissions** (natural + anthropogenic) and represents a fundamental scientific uncertainty in our understanding.

Discuss whether this/these errors could be within Annex I, RoW or natural sources, if we can. Short.

Are the errors confined to some specific sectors? This overlaps with a).

If so, this may affect the national emissions differently and thus the attribution calculations. Use expert judgements.]

## 2.4 SF<sub>6</sub>, PFCs, HFCs

(Höhne, July 2005)

### 2.4.1 Annex I emissions by country

The fluorinated greenhouse gases hydrofluorocarbons (HFCs), perfluorocarbons (PFCs) and sulfur hexafluoride (SF<sub>6</sub>) are reported to the UNFCCC by the national governments of all developed countries. They report an uncertainty of these emission estimates to be between 10% and 50%. Governments of developing countries do not have such a reporting requirement and usually do not report these emissions.

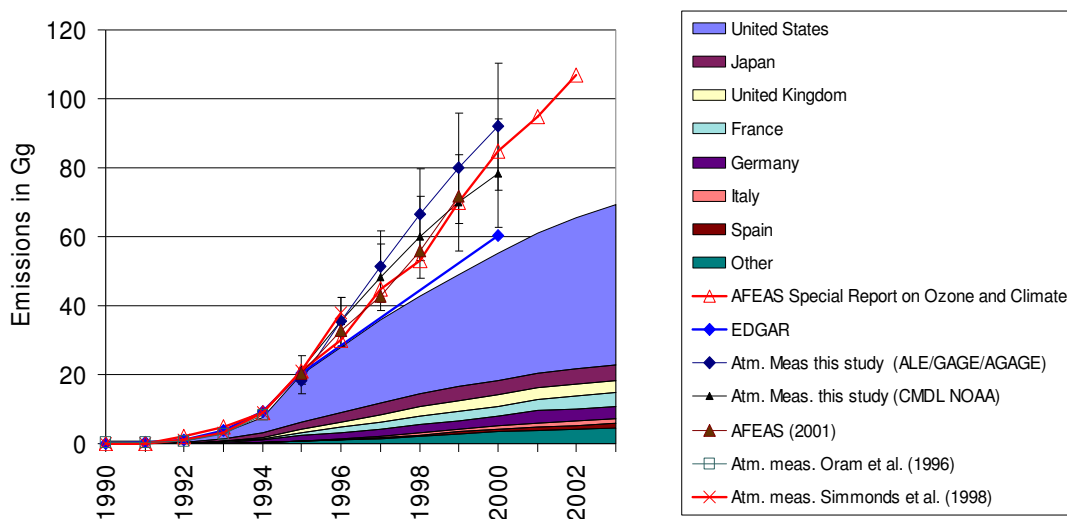
Regional emission estimates are also available from the US Environmental Protection Agency for the years 1990, 1995 and 2000 (USEPA 2002) and from the RIVM's EDGAR database for the years 1990 and 1995 (Olivier and Berdowski 2001) and for 2000 (.

### 2.4.2 Possible biases / lack of understanding of global F-gas emissions

HFCs, PFCs and SF<sub>6</sub> are (almost) completely manmade gases, have long lifetimes and their emissions can be well traced in the atmosphere. In general, the sources of these emissions are well understood. Several comparisons of the emission estimates reported by countries and other sources with estimates derived from atmospheric measurements are available (e.g. Höhne and Harnisch, 2002).

#### HFC-134a

HFC-134a was introduced as a replacement of CFCs and HCFCs and its emission started in 1990. It is increasing rapidly since. Most emissions originate from Annex I countries. The gap between reported Annex I emissions and estimates from atmospheric measurements (40% in 2003) can either be due to emissions in developing countries or the systematic underestimation of emission in Annex I countries.

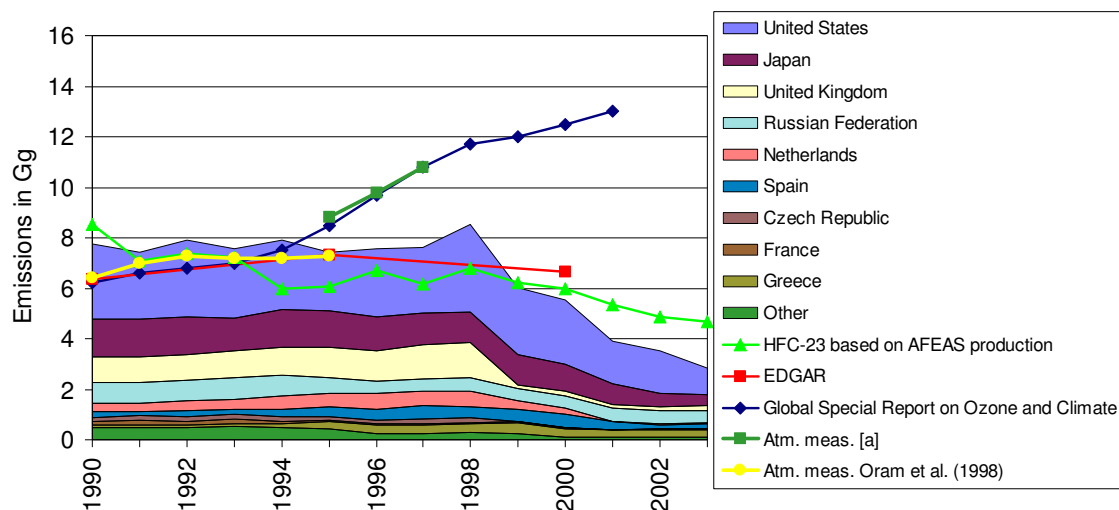


(Figure will not be in the final paper)

#### HFC-22

HFC-23 is formed as a by-product during the production of HCFC-22, which started in 1970. Depending on the conditions during the production process and whether the HFC-23 is vented, captured or destroyed, the emission rate relative to the HCFC-22 production is usually 4% to 0% by mass. It has generally been at the high end of the range prior to 1990 and declined due to increased awareness and consequently increased capture and destruction. The three major

emitters are the USA, Japan and the United Kingdom. In China, India and South Korea production has increased dramatically in recent years. For 1990, aggregate emissions of HFC-23 reported by Annex I Parties, not including developing countries, are a little higher than global emissions derived from atmospheric measurements. One possible explanation would be the use of a perhaps too high by-production factor in the emissions calculation by Annex I Parties for the early nineties. Annex I countries report that emissions have decreased substantially in the late 1990's due to the installation of a new HFC destruction system in one of its HCFC plants. This decline cannot be seen with atmospheric measurements.



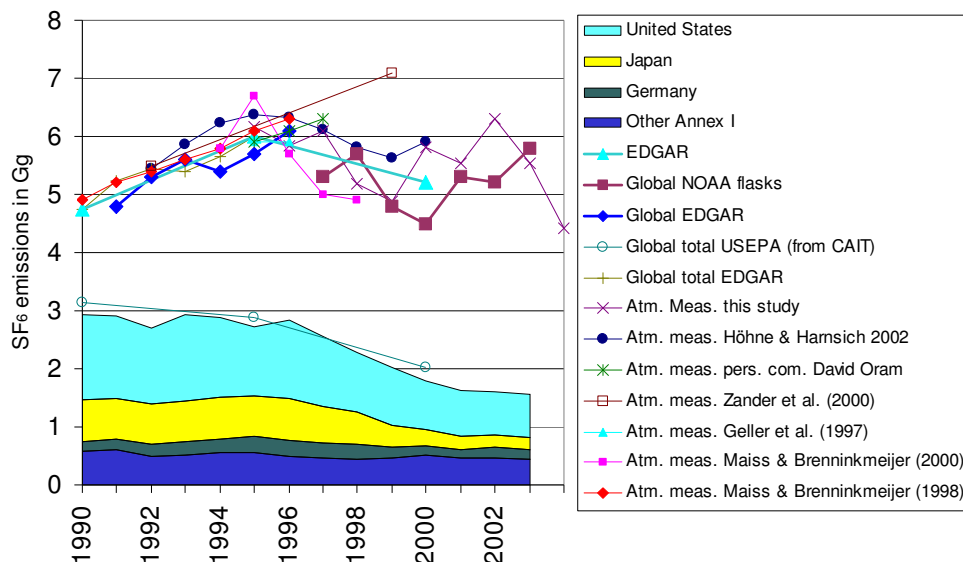
(Figure will not be in the final paper)

#### **CF<sub>4</sub> and C<sub>2</sub>F<sub>6</sub>**

CF<sub>4</sub> is mainly emitted as a by-product during the production of primary aluminium. Over the 1990's CF<sub>4</sub> has also increasingly been used in semiconductor manufacturing. C<sub>2</sub>F<sub>6</sub> is mainly used in semiconductor manufacturing but also occurs as a by-product during the production of primary aluminium (a factor 10 less than CF<sub>4</sub>). Emissions reported by Annex I countries are in broad agreement with atmospheric measurements.

#### **SF<sub>6</sub>**

SF<sub>6</sub> is used in high voltage switch gear, in aluminium and magnesium foundries and in small quantities in a number of other applications. Annex I countries report a downward trend in emissions. Magnitude and trend are almost represented by the global estimates by the USEPA. Emission estimates from atmospheric measurements using various techniques to measure SF<sub>6</sub> are in broad agreement and show much a higher emission levels: in 1995 by a factor of 2. This difference can be due to emissions in Russia and China or other developing countries but also due to a systematic underestimation of SF<sub>6</sub> emissions using too low emission factors. In addition, the downward emission trend of Annex I countries is not clearly visible from the atmospheric measurements.



(Figure will not be in the final paper)

### 2.4.3 Discussion

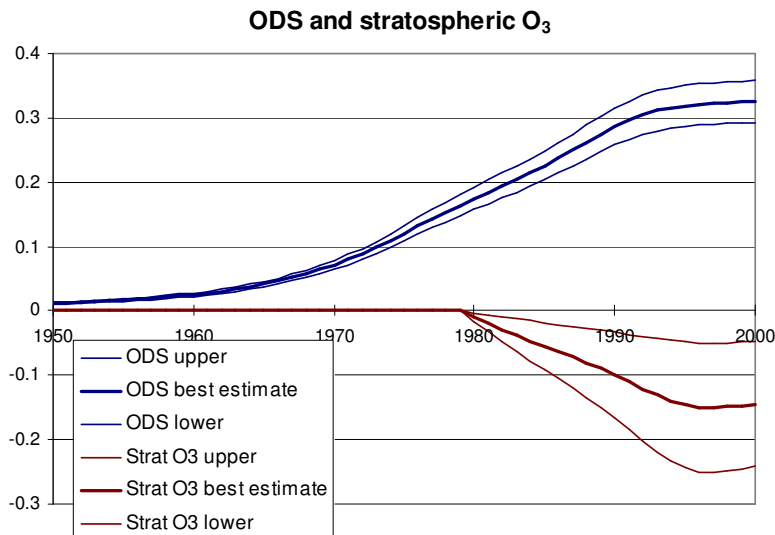
Emission estimates of the HFCs, PFCs and SF<sub>6</sub> are only in broad agreement with the atmospheric budgets. Emissions of gases slowly leaking from appliances (HFC 134a and SF<sub>6</sub>) may be underestimated, while declining process emissions may be initially overestimated. However, since emissions of these gases only have a small share compared to the other greenhouse gases, these systematic biases are likely to affect the historical contributions of countries only marginally. Total radiative forcing in 2000 were 0.0058 W/m<sup>2</sup> for HFCs, 0.0036 W/m<sup>2</sup> for PFCs and in 1998 0.002 W/m<sup>2</sup> for SF<sub>6</sub> (IPCC/TEAP 2005, IPCC 2001).

### 2.5 CFCs, HCFCs and other halocarbons

(Höhne, July 2005)

Chlorofluorocarbons (CFCs), hydrochlorofluorocarbons (HCFCs) and other chlorocarbons (CH<sub>3</sub>CCl<sub>3</sub> and CCl<sub>4</sub>) are greenhouse gases (causing direct warming) and also deplete the greenhouse gas ozone (causing indirect cooling). Their emissions have a significant contribution to radiative forcing: Direct effect in 2000 was 0.275 ± 0.025 W/m<sup>2</sup> for CFCs, 0.0328 ± 0.005 for HCFCs and 0.0155 ± 0.005 in 2000 (IPCC 2005). A history of the radiative forcing is provided in Figure 18. Their indirect effect due to ozone depletion is 0.13 ± 0.085.

The emissions of these gases are controlled by the Montreal Protocol on the Depletion of the Ozone Layer. For the purpose of this paper, emissions are not to be attributed to countries and hence are not further considered here.



**Figure 18 Radiative forcing due to halocarbons (Data source: Forster and Joshi 2005 as used in IPCC Special report on ozone and climate, figure 2.10)**

### **3 Short-lived Greenhouse Agents & other forcings**

#### **3.1 Direct aerosol forcing and uncertainties**

(Penner, Andronova, Bond, Aardenne, 15 September 2005)

The magnitude of the direct and indirect aerosol radiative forcing plays a major role in attribution of the recent and future climate change and contributes large uncertainties to estimates of the climate sensitivity (Andronova and Schlesinger, 2001). Here we examine both the historical changes in these forcings and their uncertainties.

The direct radiative forcing is derived from estimates of historical emissions. The uncertainties in those emissions are used to estimate the uncertainties in forcing, since the uncertainty in emissions is one of the most important factors in determining the uncertainty in the direct forcing the total uncertainty in estimates of the direct aerosol radiative forcing [Penner et al., 2001].

The direct aerosol forcing is associated with the scattering and absorption of sunlight by aerosols. The main aerosol constituents that contribute to this forcing are sulfate aerosols, carbonaceous aerosols (black carbon [BC] and organic matter [OM]) from fossil fuels, and smoke aerosols produced in biomass burning. The smoke aerosols from biomass burning can be further subdivided into emissions associated with biofuel burning and with open vegetation burning. Emissions of dust due to anthropogenic activities are also recognized as a potential contributor [Tegen et al., 1996], though the magnitude of the anthropogenic source is now estimated to be somewhat smaller than originally thought [Prospero??]. Nitrate aerosols formed from emissions of  $\text{NO}_x$  combining with ammonium are also recognized as a potential contributor. However, these are mainly important in future scenarios when sulfate aerosols are expected to decrease [Adams et al., 2001].

The direct radiative forcing for anthropogenic sulfate aerosols, fossil fuel organic matter and black carbon and biomass burning was estimated by Ramaswamy et al. [2001] from model

simulations. These forcings and their uncertainties (in percentage) were: sulfate aerosol:  $-0.4 \text{ Wm}^{-2}$  (100%), fossil fuel black carbon:  $0.20 \text{ Wm}^{-2}$  (100%), fossil fuel organic matter:  $-0.1 \text{ Wm}^{-2}$  (200%), biomass burning:  $-0.2 \text{ Wm}^{-2}$  (200%), mineral dust (range from  $+0.4$  to  $-0.6$ ). The uncertainties were estimated from the range of results from model simulations and “have no statistical basis”. A second estimate of the forcing and its uncertainty was based on estimates of the uncertainty associated with each factor that contributes to the direct forcing, using a simple box model [Penner et al., 2001] and assuming lognormal distribution of the uncertainties. This resulted in an estimate of  $-0.6 \pm 0.42 \text{ W/m}^2$  for the combined forcing from sulfate aerosols and fossil fuel BC and OM and its 2/3 uncertainty range and of  $-0.3 \pm 0.2 \text{ W/m}^2$  for the forcing from biomass aerosols.

To develop estimates of forcing by fossil fuel carbonaceous aerosols, we used these central estimates for forcing by fossil fuel BC and fossil fuel POM for the forcing in 2000, i.e.  $0.2 \text{ Wm}^{-2}$  and  $-0.1 \text{ Wm}^{-2}$ , respectively. Prior years forcing were estimated by scaling with the estimates of prior years' emissions. These were developed as follows.

Primary fossil fuel emissions of OM and BC were determined using the baseline fossil fuel emissions from Ito and Penner [2005] for 1985 to the present. Prior to 1985, emissions assumed the following. Between 1975 and 1985, we assumed a linear decrease with time from the emission factors for 1975 and those for 1985. Between 1970 and 1975, for developed countries, the average emission factors between those for developing countries and those for developed countries were used. Emissions between 1950 and 1970 used developing country emission factors for all countries and emissions prior to this time were scaled so that emissions smoothly connect with those in 1950. The total emissions developed in this way after 1970 are similar to those developed by Bond [private communication, 2005], but drop below hers by almost a factor of two in 1950. The OM primary emissions were multiplied by a factor of 4.3 to account for increases associated with the production of secondary fossil fuel OM [Penner et al., 2001; Cooke et al., 1999].

The 95% confidence interval associated with BC and OM emissions from fossil fuels in the year 2000 are based on the analysis of Bond et al. [2004] and are -40% to 150% and -50% to 125% for BC and OM emissions, respectively.

Forcing from biofuels used the same forcing per unit emissions as for fossil fuels, but used the biofuel emissions from Ito and Penner [2005]. Total biofuel BC emissions from Bond [private communication, 2005] are about 1.14 Tg/yr compared to only 0.77 Tg/yr from Ito and Penner [2005]. Bond's emissions increase to 1.64 Tg/yr in 1996 compared to Ito and Penner's [2005] estimates of 1.97 Tg/yr. The 95% confidence interval associated with BC and OM emissions from biofuels in the year 2000 are based on the analysis of Bond et al. [2004] and are -50% to 180% and -55% to 125% for BC and OM emissions, respectively, but clearly increase in prior years.

Forcing from open vegetation used the value from the Ramaswamy et al. [2001] for the year 2000 (i.e.  $-0.2 \text{ Wm}^{-2}$ ). Prior years' forcing was scaled to the total forcing by the open vegetation emissions from Ito and Penner [2005]. Two-sigma uncertainties for this category of emissions were estimated as  $\pm 28\%$  for the time period between 1979 and 2000 [Ito and Penner, 2005] based on an inverse modeling study, but the true uncertainties are likely larger than this estimate. Prior to this time, uncertainties are likely much larger. Emissions from open vegetation dominate the total carbonaceous emissions (Ito and Penner, 2005). Thus, the uncertainties in the total forcing were assumed to be dominated by this source. A 1-sigma value of  $\pm 30\%$  was assumed to account for factors not explicitly considered in our estimates of uncertainty. This uncertainty was increased to a value of  $\pm 60\%$  at an initial time period of 1750.

Sulfate direct forcing for the year 2000 was determined using the estimate of forcing from Ramaswamy et al. [2001] for 2000, and was scaled back in time using the estimates of emissions from Stern [2005]. Uncertainties were developed from the summary of estimates provided in van Aardenne et al. [2001]. A 1-sigma uncertainty of  $\pm 10\%$  increasing back in time to  $\pm 20\%$ .

### **3.2 Indirect aerosol forcing and uncertainties**

(Penner, Andronova, Bond, Aardenne, 15 September 2005)

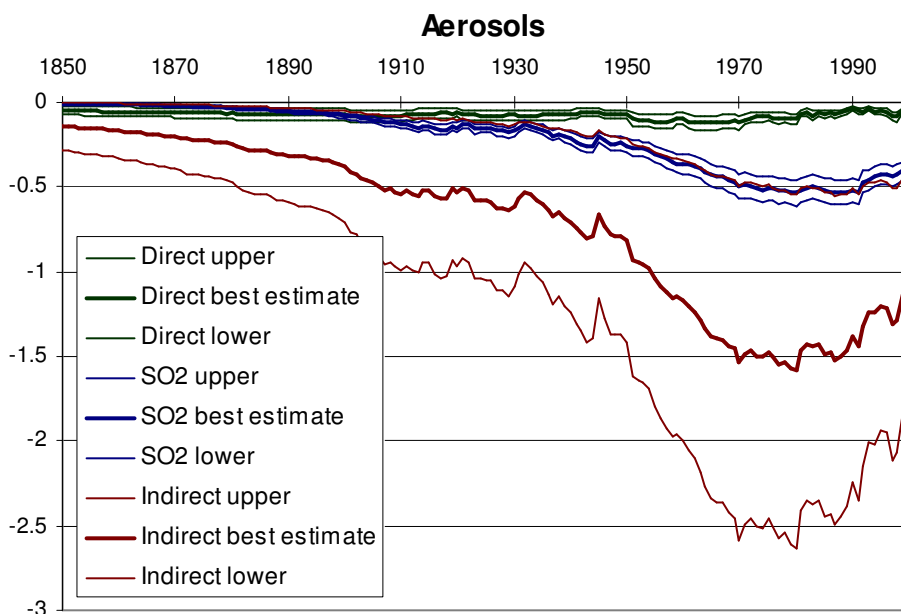
The indirect aerosol forcing, known also as “indirect effect of the aerosols” or “warm cloud aerosol indirect effect”, originates from the fact that aerosol particles change cloud properties by acting as cloud condensation nuclei and/or ice nuclei, thereby changing the optical properties of clouds. Two such effects have been recognized: (a) an effect related to a change in the droplet number concentration – the “Twomey effect”, and (b) an effect related to a change in the precipitation efficiency and cloud lifetime – the “Albrecht effect.” These effects are difficult to quantify from direct measurements, and calculation of their size is mainly derived from models of aerosol and cloud microphysics coupled to the atmospheric models (direct method), or from constraining the magnitude of the indirect aerosol forcing from the near-surface temperature observations using inverse methods and assuming that the rest components of the total forcing are known. Methodologically, the direct calculations of the aerosol indirect effect are more uncertain than estimates obtained by inverse methods (see Anderson et al, 2004), though the latter estimates actually refer to the sum total of effects not directly modelled in the study (so aerosol indirect effects on ice or changes to water vapour in the stratosphere are, for example, included). This is because in the absence of detailed observations and with the lack of necessary spatial and temporal model resolution, the direct calculation of the aerosol indirect effects has to include many “free” parameters and crude parameterizations.

The IPCC Third Assessment Report cited two estimates of the Twomey effect: the first was based on calculations by general circulation models (GCMs) and ranged between 0 and  $-2\text{W/m}^2$  in the global mean (Ramaswamy et al., 2001); the second was based on the method of propagating uncertainties using a simple box model representation of aerosol/cloud interactions on global scale and ranged between 0 and  $-2.8\text{W/m}^2$  (Penner et al, 2001). Both estimates are close to each other with the latter showing slightly wider range.

Since the last IPCC report the indirect aerosol effect has been under continuous investigation (Ghan et al., 2001; Jones et al., 2001; Lohmann and Feichter, 2001; Williams et al., 2001; Menon et al., 2002a), but with a little revisions: the Twomey effect is estimated to be between  $-0.5$  and  $-1.9\text{W/m}^2$  and the Albrecht effect is between  $-0.3$  and  $-1.4\text{W/m}^2$ . Here we will consider the forcing and uncertainties only in the first indirect effect. These uncertainties come from the following facts: (1) different definitions of the indirect aerosol forcing is used in literature (see e.g. Chuang et al. (2002), Rotstayn and Liu (2003) and Quaas et al. (2004) vs Menon et al. (2002a)); (2) the cloud albedo depends on both the cloud droplet size and the cloud thickness and cloud fraction or cover, which are difficult to predict; (3) the indirect aerosol effect depends on the background aerosol concentration, which is different in different regions. Calculation of the second indirect effect is further uncertain because there are differences in the cloud microphysics scheme used by different researchers, which is mainly related to the representation of the cloud droplet autoconversion rate.

Here we take the existing estimates from GCMs (Ramaswamy et al., 2001) as being 0 to  $-2\text{W/m}^2$ , and first assume that each value from this range is equally probable, which corresponds to a flat probability density function (pdf); second, we assume they are distributed normally with

a mean  $-1 \text{ W/m}^2$  with variance equals to  $1 \text{ W/m}^2$ . We combined these two distributions with that from the empirical estimate of Penner et al. (2001) taken here as  $-1.4 \pm 1.4 \text{ W/m}^2$  (one sigma) where we assume there is a lognormal distribution of uncertainties (Boucher and Haywood, 2001). There is not much difference in two versions of the resulting density functions: inclusion of the normal distribution instead of a flat distribution in the estimates obtained from GCMs makes the 90% confidence interval only a little narrower with difference in the median equal  $0.14 \text{ W/m}^2$ . These uncertainties were used to estimate the 1-sigma uncertainty in 2000 and scaled back in time as described below.



**Figure 19. History of radiative forcing from aerosols**

### 3.3 Tropospheric O<sub>3</sub>

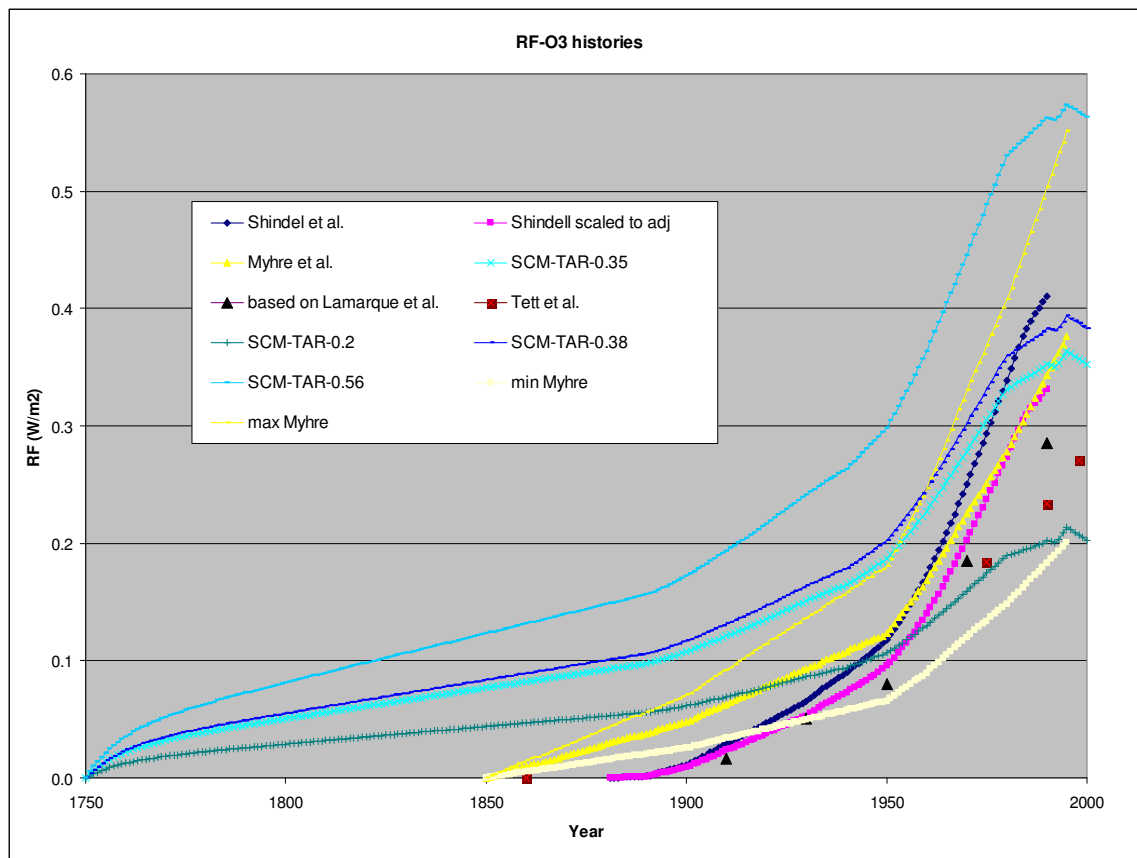
(Fuglestad, 24 October)

So far we have O<sub>3</sub> abundance/RF histories from<sup>3</sup>:

- Myhre et al. 2001 and Berntsen et al. 2000 (RF)
- Lamarque et al., 2005 (DU units)
- Shindell et al. (submitted) (RF)
- Tett et al., 2002.

In addition we have calculated the development in tropospheric ozone forcing with a SCM using the TAR expression for O<sub>3</sub> change as function of CH<sub>4</sub> concentration and emissions of NO<sub>x</sub>, CO and VOC. For the period before 1890 (when the emission database starts) we scale the increase in ozone with emissions of CO<sub>2</sub> from fossil fuels. This gives an earlier start date for tropospheric ozone increase (which needs to be discussed).

<sup>3</sup> Have asked for RF-O<sub>3</sub> from Takemura et al., JGR 2005.



**Figure 20. RF histories for tropospheric ozone based on various studies.**

Lamarque et al. (2005) gives development in ozone burden (DU). We have used the factor given by TAR for RF per change in DU to calculate a RF history for these data.

In the paper by Shindell et al. the history is given for instantaneous RF. Based on results from two different radiation transfer models (UiO/Myhre and Reading/Shine in Berntsen et al., 1997) these RF results are scaled to give adjusted RF to allow for a comparison with the other results in the figure.

### Uncertainty range

According to TAR, the global mean tropospheric  $O_3$  has increased from 25 DU to 34 DU since pre-industrial times; i.e. an increase of 9 DU. An uncertainty range of 6 to 13 DU is given. (The troposphere is defined as air with  $O_3 < 150$  ppb, see Logan (1999)).

The Dobson Unit is  $1 \text{ DU} = 2.687 \cdot 10^{16}$  molecules, globally  $1 \text{ DU} = 10.9 \text{ Tg}(O_3)$  and 1 ppb of tropospheric  $O_3 = 0.65 \text{ DU}$ . The change in  $CH_4$  alone since pre-industrial conditions would give about +4 DU global increase in tropospheric  $O_3$  alone (see TAR, Table 4.11). From Table 6.3 in TAR:  $0.042 \text{ W/m}^2$  per DU with 2/3 of models in range  $0.042 \pm 0.06 \text{ W/m}^2$  per DU.

If these uncertainties are propagated equally (rms), then  $RF = 0.38 \pm 0.18 \text{ W/m}^2$ .

This is slightly higher than the estimate given in Chapter 6 in TAR: The global average radiative forcing due to increases in tropospheric  $O_3$  since pre-industrial times is estimated to be  $+0.35 \pm 0.15 \text{ Wm}^{-2}$ .

For the RF history we suggest using the Myhre data with the uncertainty range given above.

How well does the models agree with observations?

Only a few observations and difficult to compare. Figure 21 is taken from Berntsen et al., 2000, and shows average, max and min model estimates together with observations at Pic du Midi.

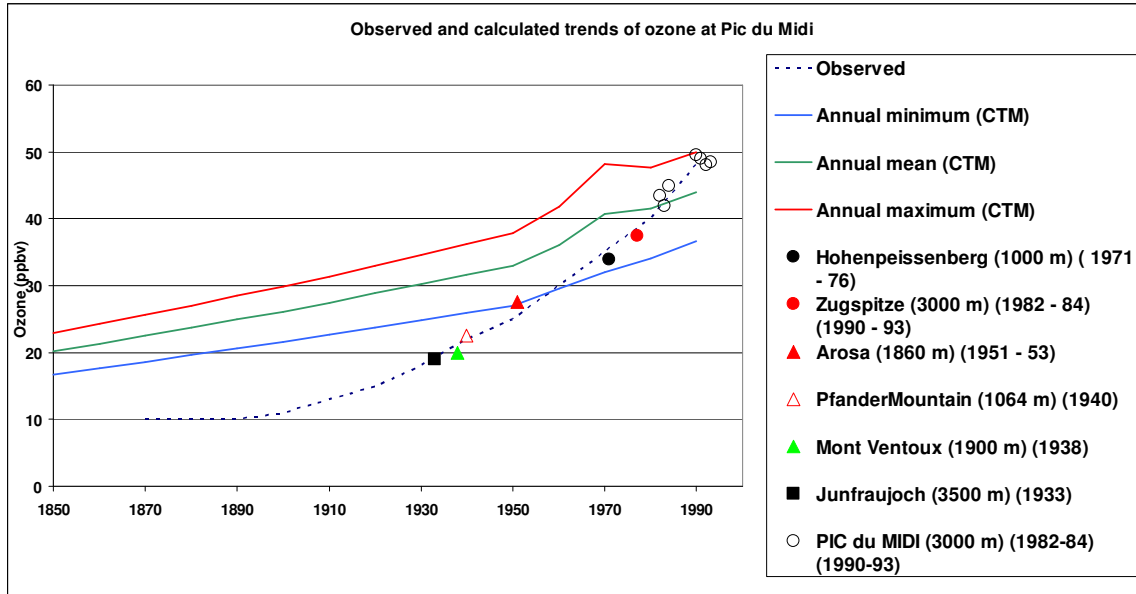


Figure 21. Observed and calculated levels of ozone at Pic du Midi.

### 3.4 Other / Natural Forcings

(Lowe, Andronova,...)

Do we want to quickly summarize here an RF range for solar and volcanic? And for LUCF-albedo change?

**Product: History of total RF from total solar irradiance changes, volcanoes, Inad-use albedo changes (cooling).**

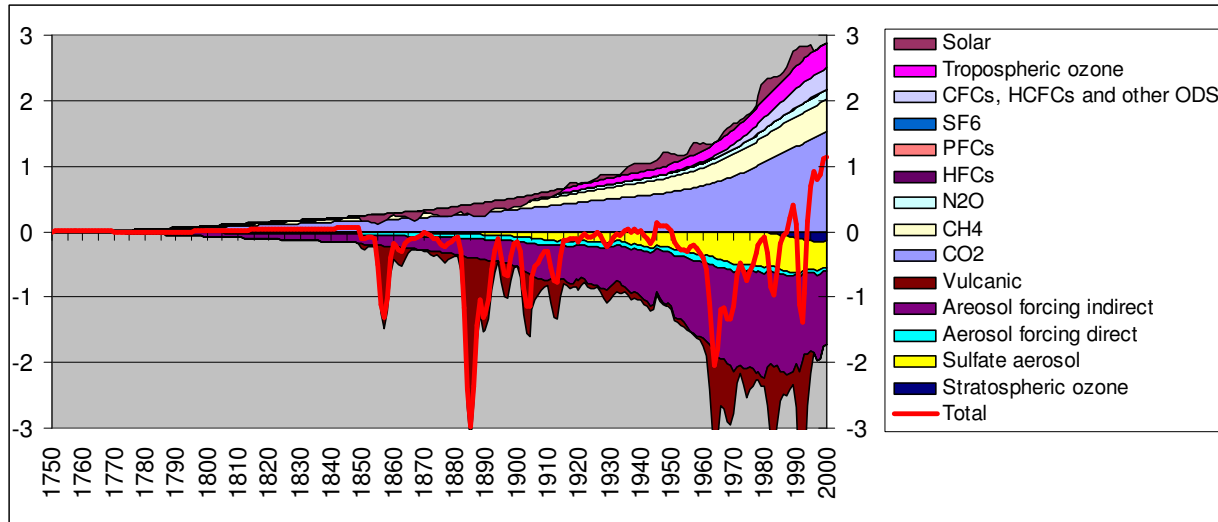


Figure 22. Total history of radiative forcing, best estimate

## 4 RF and Climate Change

(Lowe, Raper, Stott,...)

This section examines the possible errors/uncertainties in going from greenhouse gas abundances and other specified RFs to climate change. A failure/discrepancy in climate models to reproduce the last 150 years of climate change (i.e., global mean surface T and patterns, mean sea level rise) could be due to either (i) a failure of the model to simulate climate feedbacks (e.g., water & clouds) OR (ii) a possible error in the RF history. In either case, significant discrepancy reduces overall confidence in attributing climate change, even relative climate change. In case (ii), however, there are several errors that might significantly alter the relative attribution: e.g., non-linear additivity effects of different greenhouse agents, incorrect calculation of RF from abundance, missing gases (unlikely).

### 4.1 Mapping Abundances to RF

### 4.2 Reconciling RF with climate change record (forward and reverse)

### 4.3 Forward models of Annex-I emissions/ RF to climate indices

Which ones?

### 4.4 Spatial patterns and attribution from the 3 GCM runs

Product: ? Confidence that the relative RF histories are a correct measure of the relative climate change. Estimate possible errors/ systematic bias in RF.

## 5 Conclusions

...

## **Acknowledgements**

We would like to thank Wilfried Winniwarter for useful discussions....

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