NOx emissions from recovery boilers – why discrepancy between Finnish and Swedish values

In figure 1 the reported average yearly emissions from Swedish and Finnish pulp mills from year 2003 are shown in declining order as function of total pulp produced. The data is from official statistics by Finnish Metsäteollisuus (www.forestindustries.fi) and Swedish Skogsindustrierna (www.forestindustries.se).

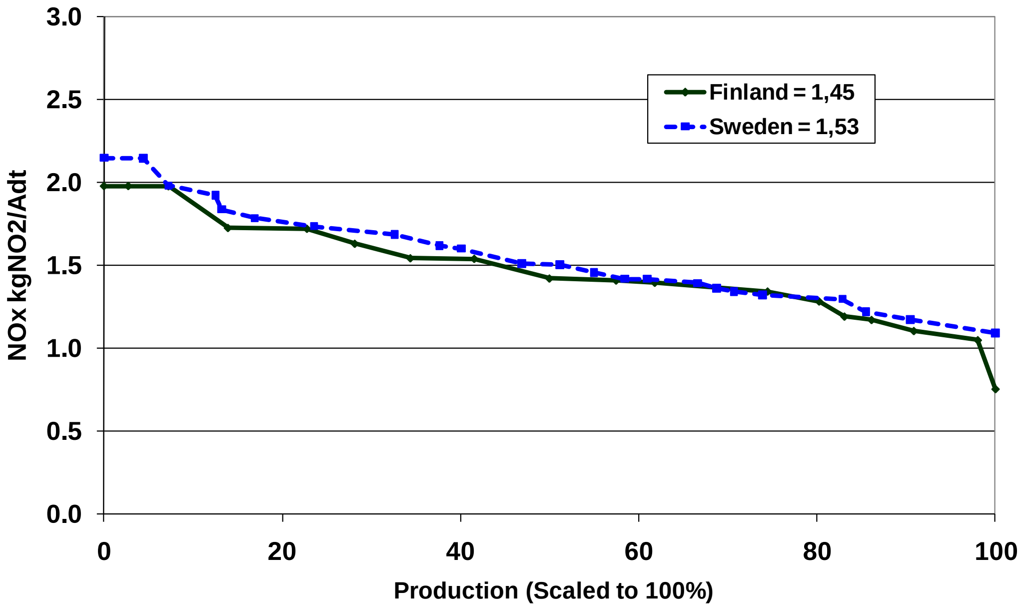

Figure 1: Average yearly emissions from Swedish and Finnish pulp mills from year 2003.

In figure 2 the reported average yearly emissions from Swedish and Finnish pulp mills from year 2008 are shown in declining order as function of total pulp produced. The data is from official statistics by Finnish Metsäteollisuus (www.forestindustries.fi) and Swedish Skogsindustrierna (www.forestindustries.se). These figures have also been reported to the EU EPER.

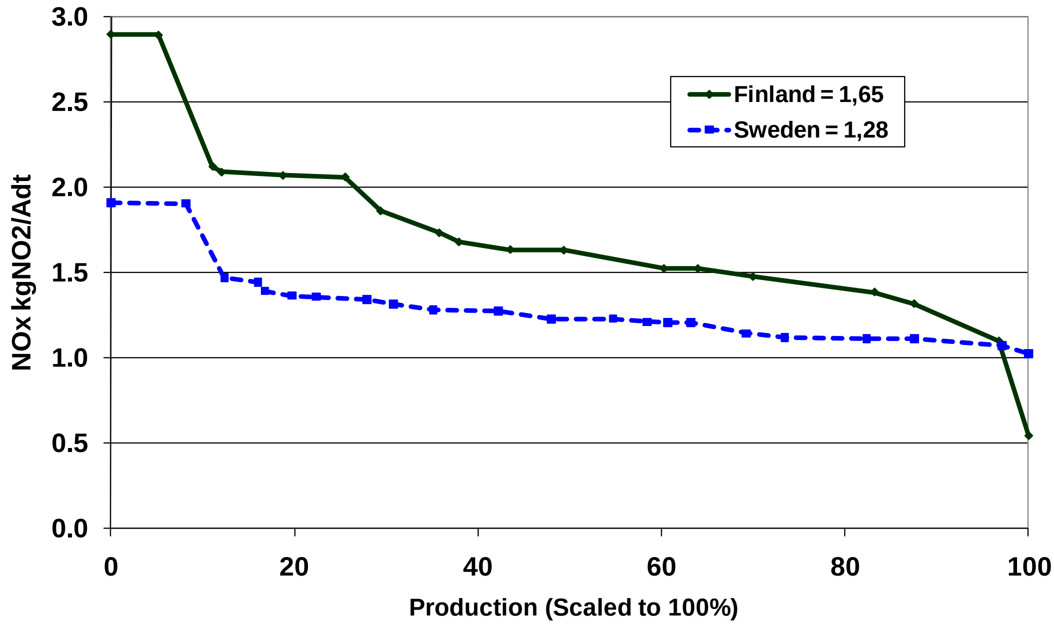

Figure 2: Average yearly emissions from Swedish and Finnish pulp mills from year 2008.

The dilemma is that in five years the average Finnish NOx emission has increased from 1.45 kg NO_2/Adt to 1.65 kg NO_2/Adt e.g. 0.2 kg NO_2/Adt. At the same time the Swedish NOx emission has decreased from 1.53 kg NO_2/Adt to 1.28 kg NO_2/Adt e.g. 0.25 kg NO_2/Adt. The difference between Sweden and Finland is now 0.37 kg NO_2/Adt.

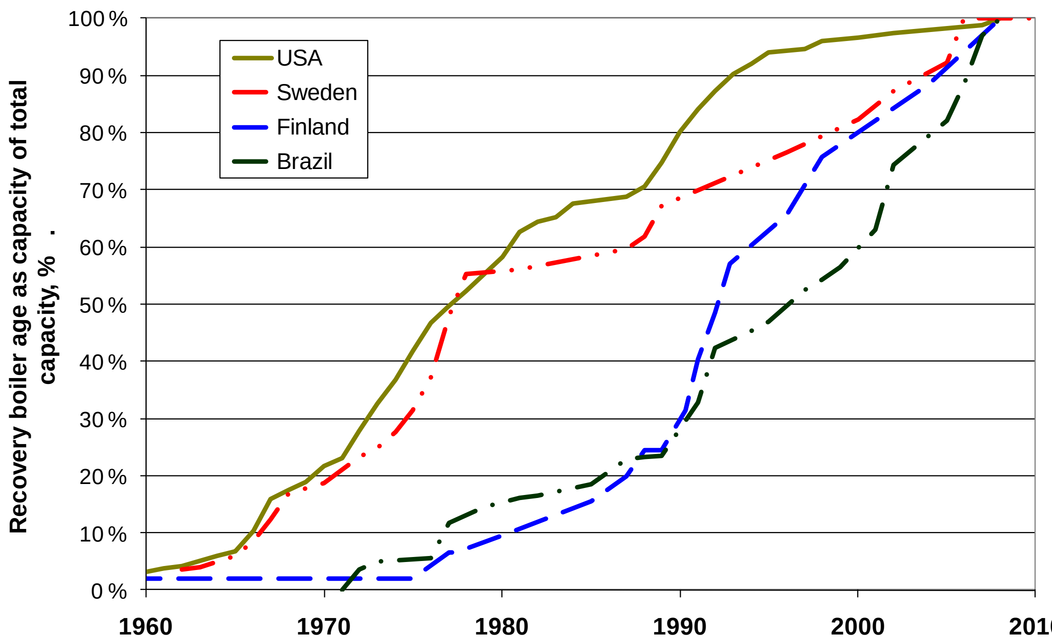

Figure 3: Ages of Brasilian, USA, Finnish and Swedish recovery boilers.

Is there a difference in Finnish and Swedish recovery boiler technology

The ages of Finnish and Swedish recovery boilers are shown in figure 3. It can be seen that Finnish recovery boilers are on average much younger than Swedish recovery boilers. Half of the Finnish boilers are younger than 20 years. Half of the Swedish recovery boilers are older than 30 years. During the period 2003 – 2008 three new recovery boilers were started in Sweden, namely Skoghall 2500 tds/d in 2005 and Östrand 3300 tds/d in 2006 and Obbola in 2007. In Finland two new recovery boilers started, namely Pietarsaari 4450 tds/d in 2004 and Kymi 4000 tds/d which started in 2008.

As far as we can state the technology base, air models and other equipment related features are similar between Finland and Sweden. The boilers in Sweden are manufactured by the same boiler vendors than in Finland. The only exception in technology is that Swedish boilers are older than Finnish.

Swedish and Finnish boilers do differ in the black liquor dry solids they fire, where DTVG is burned, where DNCG is burned, where biosludge is burned, where CNCG is burned and the liquor nitrogen content (wood species pulped).

What are the dry solids of Finnish and Swedish recovery boilers

The as fired black liquor dry solids of Finnish recovery boilers in 2004 are shown in figure 4 (ref. Suomen Soodakattilayhdistys ry, Mustalipeän polttomenetelmät Suomen soodakattiloissa, (Black liquor firing in recovery boilers) 3.5.2004, 16A0913-E0058). It can be seen that Finnish recovery boilers are on average firing close to 80 % dry solids. Dry solids increase has continued and most mills now fire 80% or above.

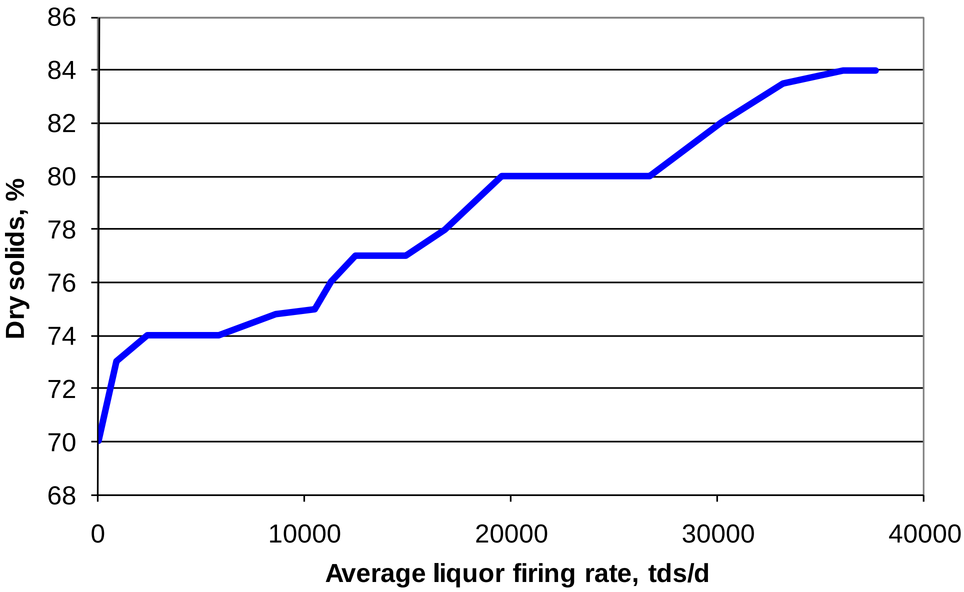

Figure 4: As fired black liquor dry solids of Finnish recovery boilers in 2004.

The as fired black liquor dry solids of Swedish recovery boilers in 2000 are shown in figure 5 (ref. Kjörk, Anders and Herstad Swärd, Solvie, 2000, Atmospheric emission of nitrogen oxide from kraft recovery boilers in Sweden (Kartläggning av NOx-utsläpp från sodapannor i Sverige). SVF-679, Technical Report, Värmeforsk, Stockholm, Sweden, Project Värmeforsk S9-807, ISSN 0282-3772, 58 p.). It can be seen that Swedish recovery boilers are on average firing close to 70 % dry solids. Swedish mills have not been investing in new evaporators. Dry solids increases have not been made and only one of the mills now fires 80% or above.

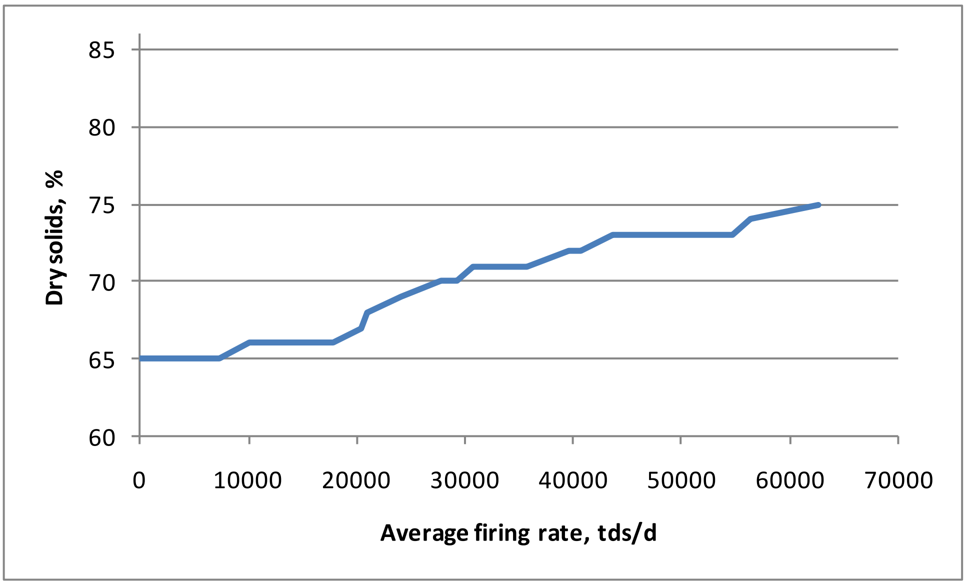

Figure 5: As fired black liquor dry solids of Swedish recovery boilers in 2000.

Our opinion is that BAT calls for 80 % or higher dry solids. Increasing dry solids increases furnace temperatures and decreases sulphur emissions, but increases NOx emissions. In Finland over 80 % dry solids is more common than in Sweden.

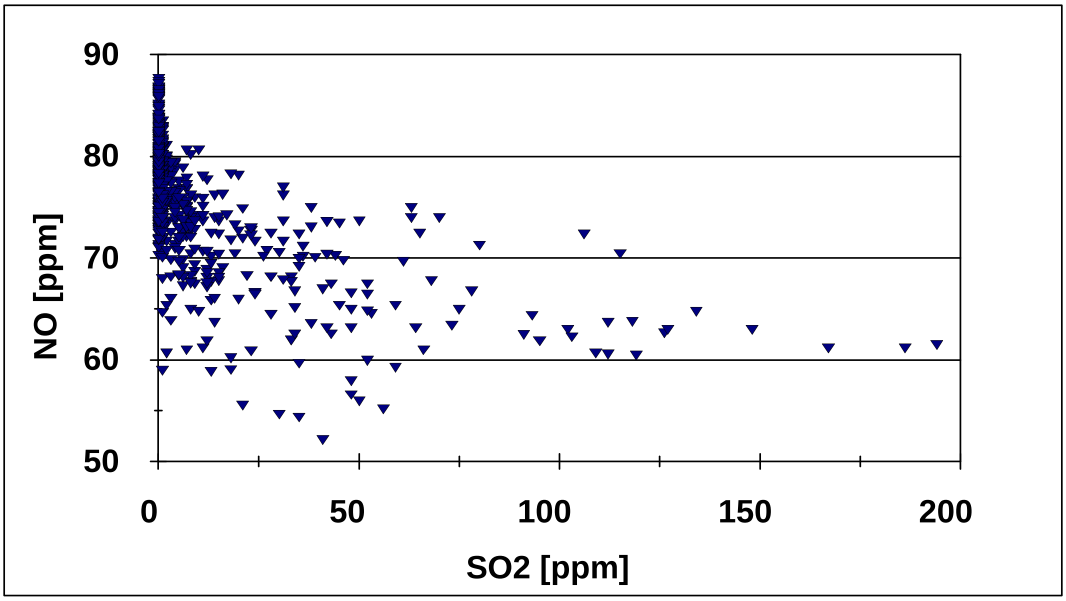

Figure 6: Correlation between SO_2 and NOx (Kaukas, UPM-Kymmene).

Cross correlation of emissions

It is widely acknowledged in all scientific literature that emissions from boilers have cross correlations. Increase in CO or sulphur containing gases has been found to decrease NOx in all kinds of boilers. In Figure 6 find results from Kaukas Finland and Figure 7 from a North American mill. If the recovery boiler operates with low SO_2, then clearly at zero SO_2 one practically loses the reduction effect of sulphur species.

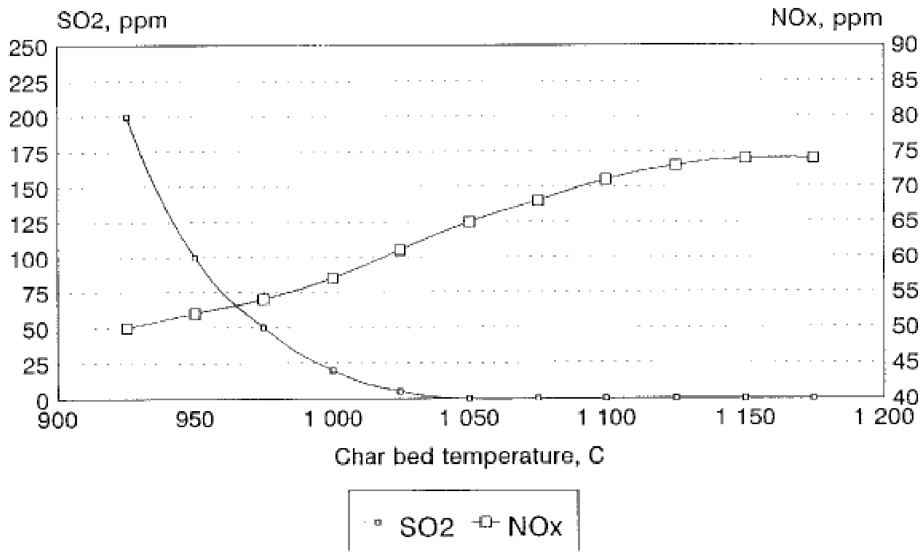

Figure 7: Correlation between char bed temperature, SO_2 and NOx in North American mill.

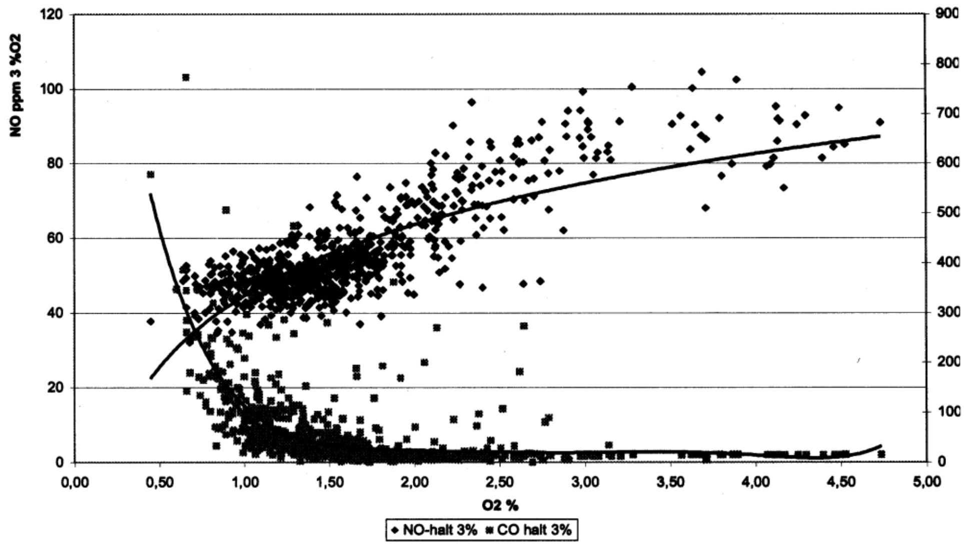

Figure 8: Correlation between O_2, CO and NOx in Swedish mill.

Figure 8 shows the effect of excess air level to NO and CO during trials at a Swedish mill. Lowering oxygen level clearly decreases NOx but increases CO.

It is typical in Finland to run recovery boiler at high furnace temperature. We try to maximize the electricity production from our boilers with high air preheat and high pressures and temperatures. Running with high furnace temperature tends to increase NOx (Figures 6 and 7).

It is the opinion of Finnish Recovery Boiler Committee, that environmentally it is better to run significantly lower CO level instead of 10 % reduction in NOx level. High CO means high amount of uncombustible, therefore the VOC and PAH (polyaromatic hydrocarbon) emissions would be higher. Finnish experience has been that even low CO means VOC and PAH emissions. These emissions are very seldom measured in recovery boilers. As far as we know no continuous measurement exists for VOC and PAH.

It is the opinion of Finnish Recovery Boiler Committee, that environmentally it is better to maximize energy production from recovery boiler. The alternative option is to produce energy with power boiler. The emissions from power boiler are not counted towards emissions from pulp mill.

Additional N-containing streams to recovery boiler

In Finland it has been more and more common to include as many N-containing streams to be combusted in the recovery boiler as possible, Figure 9. Such streams are

- vent stack gases
- strong non-condensable gases (CNGC)
- weak non-condensable gases (DNGC)
- turpentine
- methanol
- biosludge

It is clear that recycling N-containing gases to recovery boiler furnace affects the NO_2 emissions. In Sweden one of the recovery boilers burns strong non-condensable gases. Similarly only one recovery boiler burns biosludge with the black liquor.

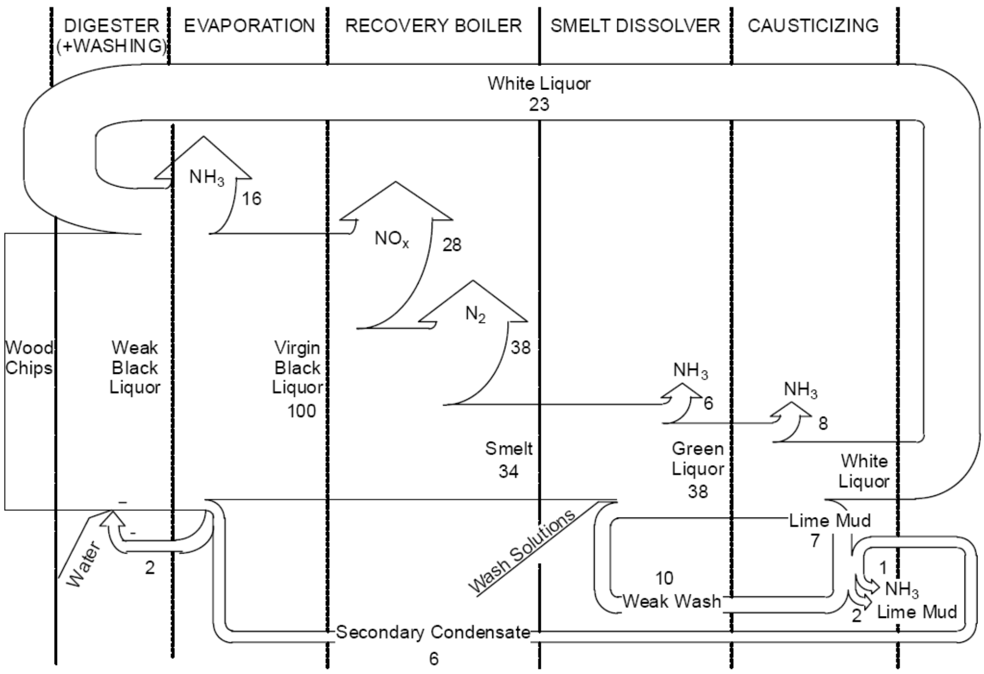

Figure 9: The nitrogen cycle in a pulp mill recovery process. The diagram is based on research at Åbo Akademi University (Kymäläinen et al.).

Pathways of Nitrogen in pulp mill

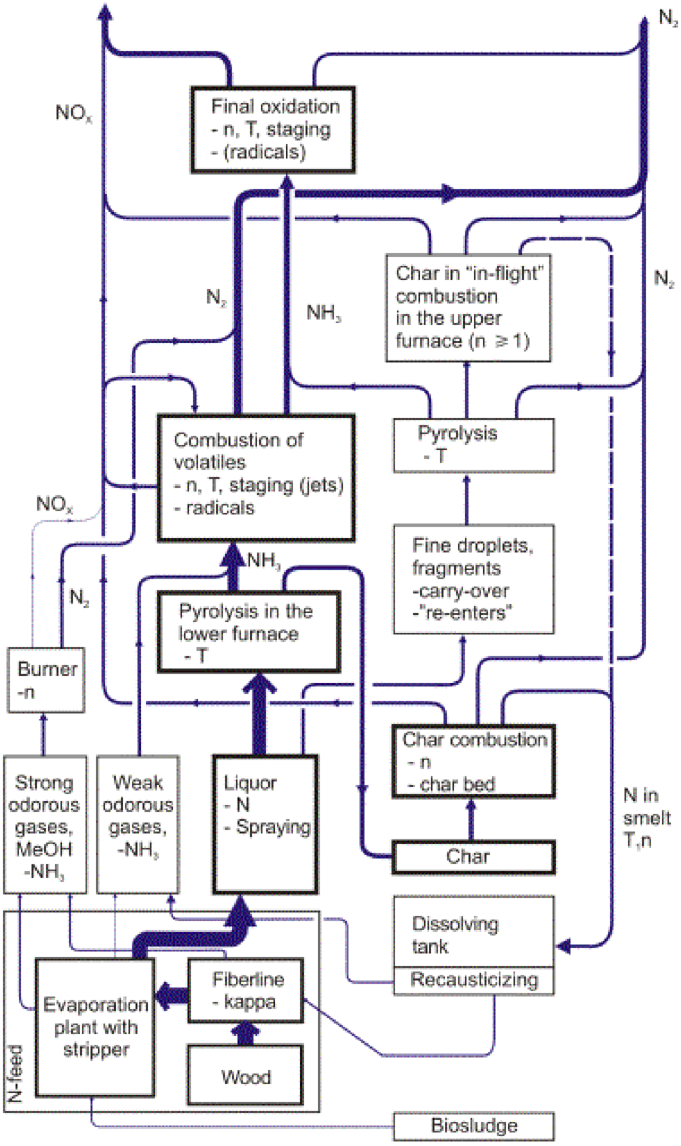

Figure 10: Nitrogen pathways in the recovery boiler.

Figure 10 shows nitrogen pathways in Recovery boiler (ref: Kari Saviharju, Kaisa Aho, 2006, Nitrogen balances in recovery boilers, TAPPI 2006 Engineering, Pulping, Environmental Conference). It should be pointed out that there are still many mills that do not e.g. collect ammonia containing flows from causticizing.

As stated in previous NOx statement to IPPC BAT process by professor Mikko Hupa, there is still much to understand of this complex picture.

Operation on different N level

Below as Figure 11 is a new technology boiler in a Scandinavian mill burning liquor that is from pulping of both Softwood and hardwood (Hardwood share about 50 %) with CNCG. Figure 12 shows a new technology boiler in Scandinavian mill burning liquor that is solely from pulping of Softwood without CNCG. Scale in both emission figures is the same.

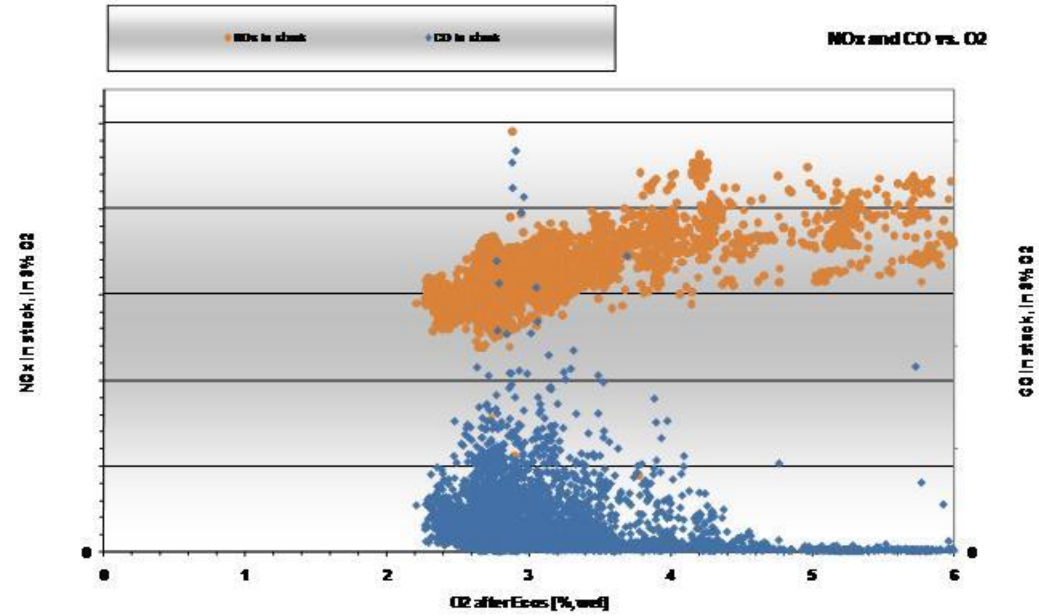

Figure 11: NOx and CO emissions burning liquor from softwood and hardwood pulping.

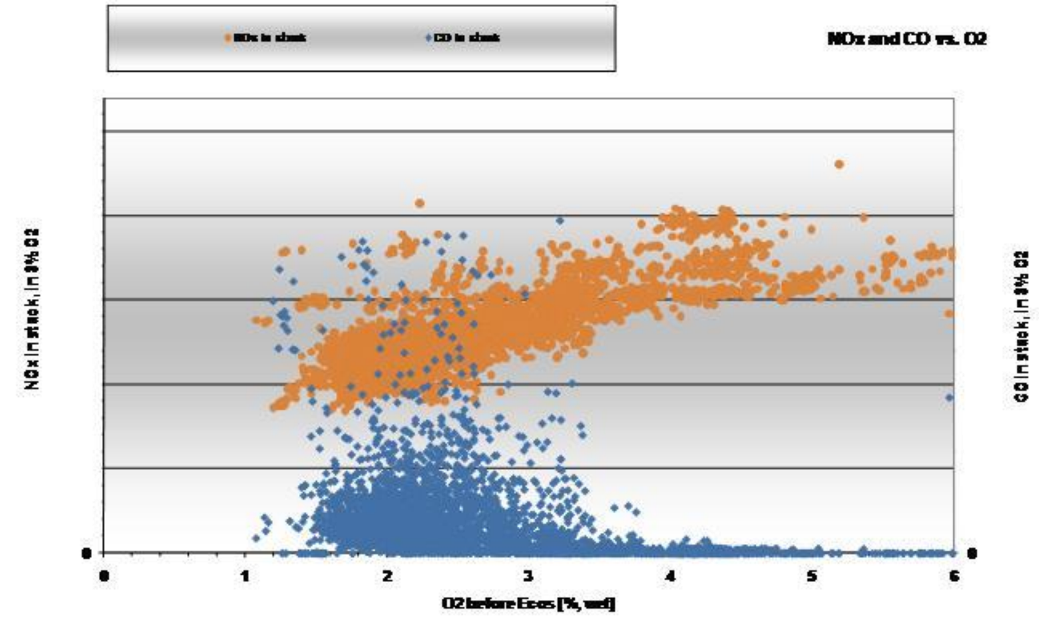

Figure 12: NOx and CO emissions burning liquor from softwood pulping.

To the best of mill and recovery boiler vendor understanding both boilers operate the same way at approximately same air pattern. It can clearly be seen that softwood liquor (N in liquor = 0.06 % KCL) results in 20 % lower emissions than softwood and hardwood liquor (N in liquor = 0.09 % KCL). The same trend has been found in laboratory tests at Åbo Akademi. Generally Sweden pulps less hardwoods than Finland. As the boilers are from the same manufacturer and represent same kind of air system and operating philosophy the role of N in black liquor is about 0.2 kg NO_2/Adt. In addition operating at ~3 % O_2 or 0.7 % higher O_2 results in another 0.1 - 0.2 kg NO_2/Adt emission.

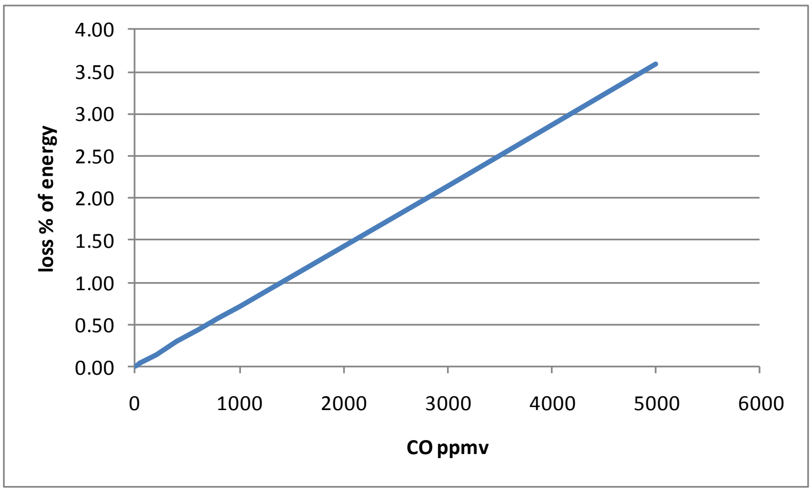

Figure 13: Effect of CO in flue gas to energy loss increase.

Losses in energy generation at recovery boiler result in higher energy generation in biomass boiler which results in higher nitrogen emission in energy generation. This nitrogen emission is not calculated to mill emissions.

Is NOx relevant emission for further reduction

NOx has not been the most important emission parameter in Finnish recovery boilers. The target as expressed through discussions with authorities has concentrated on low CO and SO_2. The reduction of NOx has been broadly evaluated, but the authorities acknowledge that NOx is not the most critical issue in Finland. Below as Figure 14, is the OMI measurement data for September 2010 for NO_2 concentration in Europe. Ozone Monitoring Instrument (OMI) is a UV-VIS imaging spectrometer contributed by The Netherlands and Finland to the NASA EOS-Aura mission. As can be seen the European pulp and paper industry does not contribute appreciably to the emissions in high NO_2 regions.

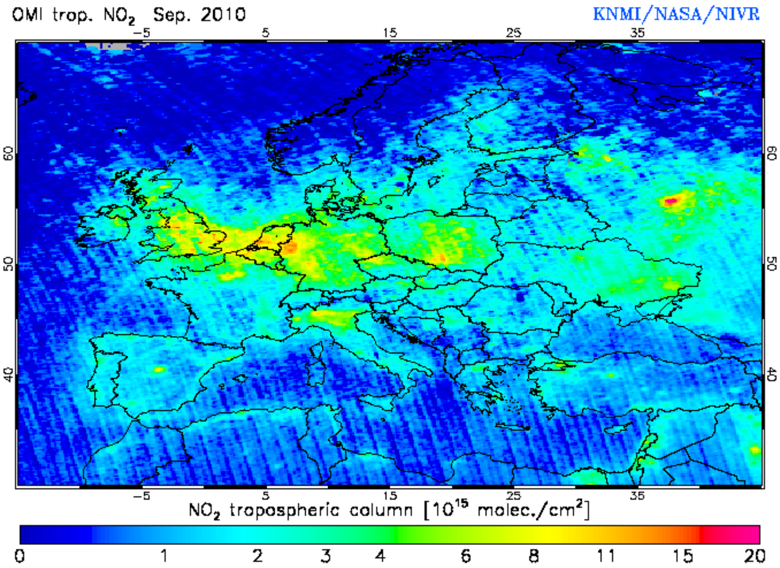

Figure 14: OMI NO_2 data for Europe in September 2010.

The NOx levels in typical Finnish mill towns are currently much lower than relevant EU regulations. Also in these mill towns as the authorities acknowledge the biggest peak comes from traffic.

Reasons for increase in Finnish NOx Emissions

It has been acknowledged that many Finnish mills have had to increase their reported NOx levels after thorough review of their reporting procedure and adoption of continuous NOx measurements.

Typically in Scandinavia the flue gas output is not measured but is calculated. These calculations have in several instances been found to result in lower values. It is clear that inherent miscalculations can result if calculated flue gas flows are used.

There are significant differences in the way Finnish and Swedish mills report their emissions based on historical practices. Finnish Recovery Boiler Committee has sought to clarify emission measurement practices between Finland and Sweden through examples and co-operation. So far we have not been able to interest our Swedish colleagues in participating.

Effect of lime kiln fuel

The NOx emissions from Sweden are reported to be lower than in Finland. In part the difference is based upon the Swedish practice of firing the lime kiln with oil. In Finland it is more common to use natural gas firing. It is widely acknowledged that oil-fired lime kilns will produce lower levels of NOx, but this does not mean that natural gas-fired lime kilns are inappropriate. Higher NOx emission levels should be allowed for lime kilns that adopt a natural gas firing system. IPPC BAT BREF for pulp and paper states that emissions from lime kilns are (ref: IPPC BAT BREF, Table 2.13 p. 43)

Nitrogen oxides (as NO_2)		
- oil firing	240-380	mg/m^3n
	130-200	mg/MJ
	0.2-0.3	kg/ADt
- gas firing	380-600	mg/m^3n
	200-320	mg/MJ
	0.3-0.4	kg/ADt

Leaving aside the broader environmental advantages of using natural gas over oil (vehicle movements, handling etc), it is more appropriate to take a holistic approach to determine the limit of NOx.

The NOx emissions from lime kilns firing biomass instead of fossil fuels are higher. This is because the nitrogen content of biomass fuels is high. Because of the burning process the nitrogen emissions cannot be reduced. Placing tight emissions will result in slower adoption of CO_2 reduction measurements. Both in Sweden and Finland some mills already use biomass firing in lime kilns, but this use will increase in the future.

Finnish Recovery Boiler Committee opinion on achieving NOx limit less than 2.0 kg/ADt

As an alternative in-stack NOx concentrations could be reduced by making substantial modifications to the operation of the recovery boiler by:

- Reducing the furnace temperature, which would result in an increase of SO_2 emissions and lower steam and electricity generation;
- Reducing the air ratio in the recovery furnace, which would result in an increase of poorly combusted emissions of CO, VOC and PAH;
- Changing the raw material from hardwood to softwood and/or
- Changing the pulping process from fine paper to cardboard

Although it is technically possible to reduce the NOx, to do so will result in an increase in other environmental impacts, like CO, VOC and PAH. It is much better to derive emission limits in a holistic way, based upon the consequences of trying to achieve those limits balanced against the actual potential impact. In this case, it is unhelpful to focus on achieving the recommended in-stack NOx concentrations if that will, as a result, cause higher emissions of carbon monoxide, VOC ,and PAH, SO_2 and TRS from the pulp mill and decrease electricity and steam production.

It can be argued that mills can choose the wood species that they pulp, but market conditions do not always favour this. It is also thought that the role of the IPPC BAT is not to require mills to utilize only a narrow range of raw materials.

The best mill in Finnish data is Kotka mills (former Laminating paper) in Kotka. Because they pulp to very high kappa they produce only close to 1000 kg black liquor /Adt . If one aims to produce fine paper then the result is over to 1600 kg black liquor /Adt. In both cases the boiler has about the same NOx concentration in the stack. But because you produce much less flue gases from 1000 kg that 1600 kg the end result is the apparent superiority of less black liquor produced. It seems absurd to us that the boiler resembling BAT in Finland would be from 1959 = the oldest still operating boiler. Similarly it seems absurd that fairly new boilers which technically are BAT should be deemed not BAT based on the mill NOx emissions.

Best practice emissions from Finnish practice are

- recovery boiler 1.2 – 1.4 kg NO_2/Adt
- lime kiln 0.3 – 0.4 kg NO_2/Adt
- gas destruction boiler 0.1 – 0.2 kg NO_2/Adt

Additionally it should be noted that coming modern practices like firing biomass or biomass based biofuels will increase lime kiln NOx emissions because all woody fuels contain more nitrogen than oil and natural gas.

Then the upper BAT emission level from pulp production should be **2.0 kg NO_2/Adt** .Since the last time BAT was determined the dry solids in recovery boilers have increased. Similarly nitrogen containing flows like DTVG, DNCG, CNCG, methanol, turpentine and biosludge are now fired in recovery boilers. Trend for biomass firing in lime kiln is gaining favour. It should be noted that none of the major manufacturers of pulp mills (=recovery equipment) is willing to guarantee low NOx emissions for the whole mill in the new European projects.

Additionally we recommend that all nitrogen containing flows (see figure 9) are counted towards the total mill nitrogen emissions i.e. mills where CNCG and DTVG are fired at recovery boiler get 0.2 kg NO_2/Adt higher emission limits than mills without.

Why Finnish mills report higher NOx Emissions than Swedish

The following facts affect mill NOx emissions

- larger share of hardwood in Finland resulting about 0.2 kg NO_2/Adt higher emission in Finland
- lower CO resulting in about 0.1 kg NO_2/Adt higher emission in Finland
- higher dry solids and hotter furnace resulting in about 0 - 0.2 kg NO_2/Adt higher emission in Finland
- firing CNCG in recovery boiler resulting in about 0 - 0.1 kg NO_2/Adt higher emission in Finland
- firing biosludge in recovery boiler resulting in about 0 - 0.1 kg NO_2/Adt higher emission in Finland
- natural gas in lime kilns 0.2 kg NO_2/Adt higher emission in Finland.

It should be outlined that all these facts do not apply to every Finnish mill. But on average these factors explain the reported 0.4 kg NO_2/Adt difference.

There are significant differences in the way Finnish and Swedish mills report their emissions based on historical practices. There should be only one way emissions in EU can be reported, not variations by country.

Added Clarification

Some of the data in the previous text has been presented without relevant sources and naming of mills in question. It is the practice of Finnish Recovery Boiler Committee not to publicise data from individual mills without written permission from each mill. If needed we can name the source mill for each figure and try to obtain permission to include the figure to the IPPC BREF document.

22.12.2010 Finnish Recovery Boiler Committee

The Finnish Recovery Boiler Committee has promoted safe, economic and environmentally friendly operation of recovery boilers and closely related processes since 1964. The members of the Committee include pulp mills, recovery boiler manufacturers, a number of insurance companies and automation system suppliers, engineering companies and research organisations in Finland.