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Mercury exposure in Dutch households supplied by Groningen gas

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BACKGROUND

In the last decades mercury has been identified as a hazardous substance and the society has become conscious of its associated risks. As a result, regulation has been put in place to abolish or minimize the use of mercury in consumer products. In addition, intervention values for working (Treshold Limit Values, TLV) and living conditions have been established by the relevant authorities.

Natural gas contains mercury, although it may be below the detectable limits depending on methodology used. A substantial part of the mercury originally present in the gas is removed through processing the natural gas (removal of fluids). The efficiency of this process strongly depends on the method and conditions.

Monitoring programs executed by Gasunie Transport Services (GTS) and NAM have shown that there is a great variety in the mercury content of natural gas. Of those fields operated by NAM, the highest concentration of mercury in *unprocessed* gas is in the Groningen field. *Processed* Groningen gas is supplied to domestic users in The Netherlands and abroad.

The monitoring programs assessed the entire gas production and GTS transport chain. Until recently, mercury levels observed at GTS gas receiving stations were only a fraction of the mercury delivered at the entry points. This leads to the conclusion that mercury was absorbed in the GTS pipeline system. A KEMA study in 2012 showed that the mercury levels in gas in some of the exit points of the GTS transportation system had increased over time to values up to 10 µg/ m^3. Higher values could suggest that little to no adsorption takes place in those pipelines, whereas significant reduction in mercury level is still measured over other parts of the pipeline system.

In 2013, GTS and NAM requested a *sample* test to be conducted by KEMA to determine the mercury concentration in gas that is supplied to domestic users (also referred to as households)[1]. The findings show that mercury is present at the gas entry points of households, with concentrations between 0.32 µg/m^3 and 0.75 µg/m^3, with one outlier of 6.9 µg/m^3. This implies that consumers could be exposed to mercury by combustion of the gas which could affect the indoor air quality.

It is key for NAM and GTS to understand the relation between mercury in indoor air and mercury in the supplied gas to assess the risk of health exposure of Dutch households.

AIM

The purpose of this study is twofold:

1) Develop a model based on widely accepted parameters to assess the exposure of Dutch households to mercury in air through gas combustion.
2) Determine if current concentrations of mercury in natural gas can lead to exposure levels above the lifetime exposure limit of 0.05 µg/m^3 in indoor air set by RIVM (Dutch national authority for Public Health and Environment).

This study will address the following aspects:

1. Current health exposure for different types of Dutch households
    a. Day average exposure in µg/m^3 indoor air
    b. Peak exposure pattern in µg/m^3 indoor air
2. Maximum mercury content in gas based on RIVM lifetime exposure limit
    a. Average daily exposure equal to 0.05 µg/m^3
    b. Exposure to peaks > 0.05 µg/m^3

METHODOLOGY

This study is designed by using references to existing default values of RIVM for the key variables. The key variables, and references to RIVM, are described in the following sections.

<u>Reference to RIVM mercury limit</u>[2]

RIVM has defined a lifetime exposure limit for mercury in indoor air at 0.05 µg/m^3. This is based on the assessment of an average annual exposure for all groups, including children and pregnant women.

The same document advises a short duration (1 week) exposure limit of 10 µg/m^3. This limit is not considered applicable for the daily exposure of mercury in air due to semi continuous gas consumption in households. This exposure limit relates to mercury spills through accidents (e.g. mercury spills through broken mercury temperature meter). Such spills are considered outside the scope of this study.

[1] KEMA Rapport september 2013 'Inventarisatie kwikgehaltes bij eindverbruikers'
[2] RIVM (2011), GGD-richtlijn medische milieukunde, kwik in het binnenmilieu en gezondheid, RIVM rapport 609300021/2011

The RIVM lifetime exposure limit is based on a recommended exposure of the European Commission. However, since its inception in 2001, this value has not been adopted in EU regulations. Multiple health-based recommended limits for mercury in air exist (a range from 0.05 $\mu g/m^3$ to 1 $\mu g/m^3$), published by different organizations. Because the scope of this study only includes Dutch househouds, the RIVM lifetime exposure limit of 0.05 $\mu g/m^3$ is used.

Key variables of exposure

- Sources of gas combustion affecting indoor air quality: 4-burner stove (cooking) and water boiler (heating water)
    - Please note that a water boiler (also referred to as water heater or geyser, in Dutch: geiser) refers to a water heating device that is placed in the cooking/living area which emits its combustion air to the indoor air. Different regulations apply for those houses build after the year 2012. Bouwbesluit 2012[3] dictates that water boilers need to have a combustion air removal channel connect to outside air, and thus becomes a closed system which does not emit to indoor air. However, this is only a requirement for newly build houses.
- Amount of gas consumption
- Duration in which the gas is released and combusted in the room (also referred to as the consumption pattern)
- Ventilation of the room where gas is consumed
- Mercury content of the supplied gas

The following variables are considered not significantly affecting the model:

- Sources of gas: Heating, a closed system and thus the gas combustion does not affect the indoor air quality
- Number of people in a household

The following variable is considered constant:

- Ventilation considered to take place at a constant rate (not intermittent)

[3] Ministerie van Binnenlandse zaken en Koninkrijk relaties (2012) Bouwbesluit 2012; Toelichting toevoer van verbrandingslucht en afvoer van rookgas.
http://www.google.nl/url?sa=t&rct=j&q=&esrc=s&frm=1&source=web&cd=1&ved=0CCoQFjAA&url=http%3A%2F%2Fwww.rijksoverheid.nl%2Fbestanden%2Fdocumenten-en-publicaties%2Fbrochures%2F2012%2F06%2F11%2Finfoblad-toevoer-van-verbrandingslucht-en-afvoer-van-rookgas-bouwbesluit-2012%2Finfoblad-toevoer-van-verbrandingslucht-en-afvoer-van-rookgas-bouwbesluit-2012.pdf&ei=UJpCUpX7FlnX0QXRvYGAAQ&usg=AFQjCNGolsShjaEopYAUA8DlDfkcWFRf1g&bvm=bv.53077864,d.d2k

Situations described in model: Type of household and scenarios

Concentration of mercury indoor air will be modeled by using three types of housing (A, B,C) which represent the different circumstances of accommodation.

Type of household	Definition	Volume of space [m^3]	Total gas consumption[4] [m^3/yr]	Gas consumption for cooking and heating water[5] [6] [m^3/yr]
House A	Small shared student kitchen	15	900	440
House B	Family house with large kitchen	60	1590	440
House C	Detached house	120	2220	440

For each type of house, two ventilation scenarios will be calculated to represent a range of housing conditions representing variations of housing (degree of insulation, new build or old etc). The ventilation variable - a key parameter - is described in the next chapter.

Assumptions

The model is based on variables, which each are assigned a value for calculations purposes. The values chosen are those that are publicly available or default values advised by RIVM. The variables and their respective values are described below.

Ventilation

Ventilation strongly depends on the housing conditions; including presence of windows, degree of insulation, or presence of mechanical ventilation. Therefore, the model represents the variations in ventilation circumstances by means of range. The worst case scenario is based on a ‘realistic worst case scenario’ as defined by RIVM and a ‘best case scenario’ in which ventilation factors are used as described in NEN1087 (Bouwbesluit).

VENTILATION (turnover/hour)	House A	House B	House C	
Realistic worst case scenario	2,5*	0,6*	0,6*	RIVM default values for ventilation[7]
Best case scenario	5	1.7	1.3	Bouwbesluit, NEN1087[8]

* RIVM provides default values for ventilation in different rooms of house which represent a realistic worst case scenario. Based on various studies, a 25th percentile is estimated for ventilation rates. When combined with other 25th percentile default values for other parameters including room size, the RIVM considers the calculated exposure to be in the 99th percentile (RIVM, 2006, page 11).

[4] Nuon (2013) Gas consumption domestic households: http://www.nuon.nl/energie-besparen/e-manager/gemiddeld-energieverbruik.jsp

[5] ENECO (2011) Gas consumption for cooking and using a water boiler: http://thuis.eneco.nl/~/media/eol/pdf/tarieven-2011/tarieven-variabel-onbepaalde-tijd/gas-per-1-januari-2011

[6] Personal Communication 5.1.2.e (July 2013) with ATAG-Pelgrim

[7] RIVM (2006) General Fact Sheet: Limiting conditions and reliability, ventilation, room size, body surface area. Updated version for ConsExpo 4. Authors: H.J.Bremmer, L.C.H. Prud’Homme de Lodder, J.G.M. Van Engelen.
Please read table 7 ‘Defaults for ventilation rates in domestic homes’ on page 19 of 31.

[8] NEN1087 (Bouwbesluit 2003, 2012)

The best case scenarios are based on the following guidelines provided by Bouwbesluit 2003 and 2012, NEN1087:

Ruimte	Ventilatie
15 m^3	75 m^3/hr
60 m^3	100 m^3/hr
120 m^3	150 m^3/hr

This study has chosen two scenarios which represent a range of turnovers per hour. This range of minimum to maximum is to show the sensitivity of the mercury concentration relative to the ventilation circumstances for different types of houses. Houses build before 2003 could either have slightly more ventilation (poor insulation in older houses) or less (houses which have been well insulated but no mechanical ventilation).

A ventilation factor of 2.5 implies that the total volumetric content of the room is replaced two and a half time within the hour with clean air. It is assumed that, with the replacement of air, no mercury remains behind. However, a certain amount of mercury in air, also referred to as a background value, is accounted for.

Gas consumption

For the gas consumption for cooking 65 m^3/year is used as established by ENECO[9, 8]. This number is similar to industry information from ATAG-Pelgrim (personal communication, July 2013). ATAG-Pelgrim states that a general 4-burner stove uses a maximum of 2 m^3/hr. Preparing a dinner will take about 20 minutes with a capacity of 25% of the stove. This results in a gas consumption of 60 m^3/year, and thus similar to ENECO number of 65 m^3/year.

Gas consumption for cooking and water boiler is defined by ENECO at 440 m^3/year, which means a gas consumption for a water boiler of 375 m^3/year. According to information from ATAG-Pelgrim (personal communication), a general water boiler uses 1.3 m^3/hour. To achieve a gas consumption of 375 m^3/year, the water boiler must be used for 1 hour each day. This is considered realistic. In all scenarios the average gas consumption reference of ENECO of 440 m^3/year will be used.

Gas consumption for heating the house is considered to have no impact on indoor air quality since the flue gasses are discharged via exhaust pipes. It is assumed that the gas consumption for cooking and water boiling are independent from the size of the house.

Gas consumption pattern

The exposure of individuals depends both on how much gas is used (in this model assumed constant 440 m^3/year) and the pattern of use (continuous, once a day or several moments during the day). All three patterns are considered and in our view the worst case scenario depicted in the graphs below.

Mercury concentration in gas

For the purpose of this exercise a value of 20 µg/m^3 is used for mercury in gas delivered to end consumers.

[9] ENECO (2011) Gas consumption for cooking and using a water boiler: http://thuis.eneco.nl/~/media/eol/pdf/tarieven-2011/tarieven-variabel-onbepaalde-tijd/gas-per-1-januari-2011

Background value

A certain amount of mercury is constantly present in air. RIVM considers the background value of mercury in air in the Netherlands to be 3 ng/m^3 [10].

Homogenous diversion of mercury in air

In this study it is assumed that indoor air quality, and the thus the mercury concentration, is homogenous in air due to ventilation and air circulation. In practice, higher mercury concentrations could be found near the emission point.

Oxygen content in air

Combustion of gas in a poor ventilated small room leads to lowering the oxygen content. The minimum oxygen content is 17% oxygen in air[11,12], any lower values of oxygen will result in adverse health effects. This means that the gas combustion has to stop when the oxygen concentration becomes below 17%.

Carbon monoxide in indoor air

In this study it is assumed that indoor air quality is not affected by carbon monoxide. Carbon monoxide is formed when the combustion is incomplete. Incomplete combustion may occur as a result of oxygen deficiency. This exposure model considers the oxygen content to be equal to or above 17%. Since any lower values of oxygen result in immediate non-mercury related health effects. Another cause for incomplete combustion can be a faulty stove or water boiler, which is out of the scope of this study.

Location of individuals continuously in exposure area

This study is based on the assumption that the individuals remain in the same space continuously. This assumption implies 100 % exposure to the increased mercury in air as a result of combustion of gas. It is argued that the movement of individuals in and out the increased mercury exposed area is a sensitivity of a model that is too complex to model and allows for various interpretations which would not contribute to the validity of this model.

Other sources of mercury in the household affecting the indoor air quality

Indoor air quality can be affected by multi mercury sources, including broken mercury barometers, mercury temperature measurements or LED- lights. To measure the impact of these mercury incidents is very difficult due to the wide range of chance and impact predictions which vary per case. Therefore, it is not included in this exposure model which focuses on the exposure impact as a result of mercury in the gas consumed by the household.

[10] RIVM (2011), GGD-richtlijn medische milieukunde, kwik in het binnenmilieu en gezondheid, RIVM rapport 609300021/2011
[11] RIVM (2006) SIR adviesrapport 10223A00. De veiligheids- en gezondheidseffecten van werken in een besloten hypoxische omgeving. http://docs.minszw.nl/pdf/92/2008/92_2008_1_19676.pdf

[12] Gasunie (1980) Physical properties of natural gases.

PART 1 RESULTS – Current health exposure households

A. Average exposure

The concentrations µg/m^3 below show the exposure of Dutch households for three different types of houses, and accounting for two ventilation scenarios. All of the values are below the 0.05 µg/m^3 limit of RIVM, with the highest exposure value being 0.030 µg/m^3.

	Realistic worst case [µg/m³]		Best case [µg/m³]	
	Ventilation	Hg concentration	Ventilation	Hg concentration
House A	2.5	0.030	5	0.016
House B	0.6	0.030	1.7	0.013
House C	0.6	0.017	1.3	0.0097

The average exposure values have been calculated using the following formula:

$$average\ concentration\ (\mu g/m^3) = \frac{gas\ consumption\ (m^3/hr) \times mercury\ conc.\ in\ gas\ (\mu g/m^3)}{ventilation\ rate\ (m^3/hr)} + background\ (\mu g/m^3)$$

B. Peak exposure in worst case scenario

The aim of this section of the study is to test whether one could measure concentration values above 0.05 µg/ m^3 if a short (e.g. 15 min) test on indoor air quality would be conducted. The RIVM advises the GGD, the Municipal Health authority in the Netherlands (GGD, Gemeentelijke gezondheidsdienst) on indoor air quality. If the GGD finds indoor air quality containing > 0.05 µg/ m^3 of mercury, they are advised by the GGD to check the emission sources of mercury in that particular home.

PLEASE NOTE that the RIVM does not regard a peak exposure > 0.05 µg/m^3 as harmful, as long as the average lifetime exposure remains < 0.05 µg/m^3.

The worst case scenario is based on a 'realistic worst case scenario' as defined by RIVM[13]:

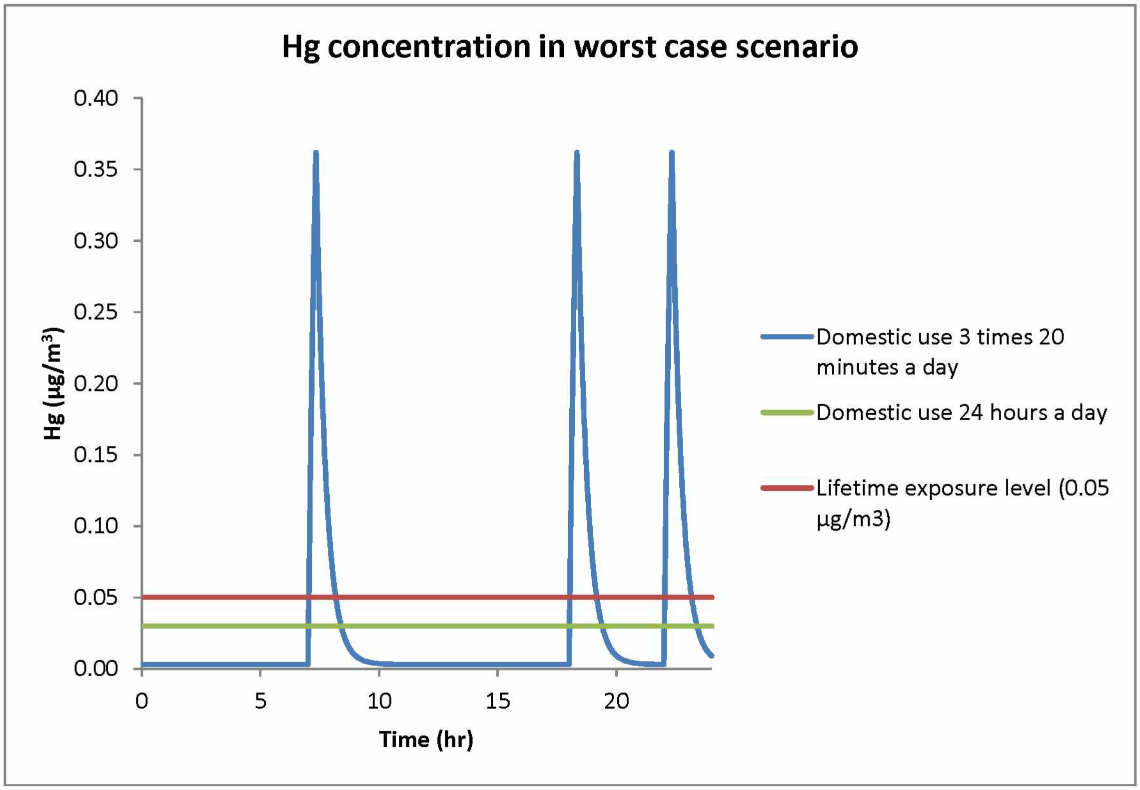
- Kitchen size: 15 m^3 (house A)
- Ventilation: 2.5 turnovers/hour

The characteristics of peaks in mercury concentrations in indoor air in the worst case scenario depend on the gas consumption pattern of a household. Gas combustion requires oxygen. A lack of oxygen therefore limits the combustion, and thus the peak of mercury. If the total daily gas consumption of 440 m3/year (1.2m3/day) were consumed in only one hour, the gas combustion has to stop after 25 minutes due to decreasing oxygen content in the room. The mercury concentration in air for the room will become 0.42 µg/m^3. For this gas consumption basis, the mercury concentration would be above 0.05 µg/m3 for 1.5 hour. This is also visualized in the graph shown in Appendix I.

[13] RIVM (2006) General Fact Sheet: Limiting conditions and reliability, ventilation, room size, body surface area. Updated version for ConsExpo 4. Authors: H.J.Bremmer, L.C.H. Prud'Homme de Lodder, J.G.M. Van Engelen.
Please read table 7 'Defaults for ventilation rates in domestic homes' on page 19

It is more realistic to assume that the total daily gas consumption of 440m3/year is used at 3 different times throughout the day for 20 minutes at a time (at 7:00 am, 6:00 pm and 10:00 pm). For this gas consumption basis, the mercury concentration would increase to 0.37 µg/m^3 and remains above 0.05 µg/m^3 for an hour each time.

The results for the different gas consumption patterns and the consequent mercury exposure are shown in graph 1.



Graph 1: Mercury concentration in a room of 15 m^3 for different gas consumption patterns with a ventilation of 2.5 turnovers/hour.

The above stated results indicate that – despite the daily average exposure being below the recommended 0.05 µg/m^3 limit of RIVM - one could measure mercury concentrations above 0.05 µg/m^3. This would only occur if a test is conducted for a short period of time (< 3 hours).

PART 2 RESULTS – Maximum mercury concentration in gas not to exceed RIVM level

A. Maximum mercury concentration in gas for to prevent life time exposure limit exposure of 0.05 µg/m^3 mercury in indoor air

As mentioned in part 1, an average gas consumption of 440 m^3/year results in a mercury concentration in the 'worst case scenario' of 0.030 µg/m^3. If one were to reach the lifetime exposure limit of 0.05 µg/m^3 - using 440 m^3/yr of gas - the mercury concentration in supplied gas would be equal to 35 µg/m^3.

B. Maximum mercury concentration in to prevent any measurable peak above 0.05 µg/m^3 in indoor air

To prevent the presence of mercury peaks > 0.05 µg/m^3 in indoor air, the following concentrations of mercury in Groningen gas apply.

	Realistic worst case (µg/m^3)			Best case (µg/m^3)		
	Ventilation	1 hour	20 min	Ventilation	1 hour	20 min
House A	2.5	2.25*	2.60	5	2.94	3.60
House B	0.6	3.15	7.70	1.7	4.80	9.20
House C	0.6	6.20	15.5	1.3	8.20	17.0

* Due to decreasing oxygen content in room, gas combustion has to stop after 25 minutes

PART 3 RESULTS – Current health exposure households based on KEMA sampling results

A. Average exposure

The concentrations µg/m^3 below show the average exposure of Dutch households for three different types of houses, and accounting for a range of ventilation. The mercury concentration in supplied gas is assumed to be 6.9 µg/m^3, which is the highest concentration measured during the KEMA sampling program.

	Realistic worst case (µg/m^3)		Best case (µg/m^3)	
	Ventilation	Hg concentration	Ventilation	Hg concentration
House A	2.5	0.012	5	0.008
House B	0.6	0.012	1.7	0.006
House C	0.6	0.008	1.3	0.005

B. Peak exposure in worst case scenario

When the total daily gas consumption of 440m3/year is used at 3 different times throughout the day for 20 minutes at a time (at 7:00 am, 6:00 pm and 10:00 pm), the mercury concentration would increase to 0.13 µg/m^3, and remains above 0.05 µg/m^3 for 40 minutes each time.

CONCLUSION

Based on the outcome of the model, one can conclude that the mercury concentrations in gas for domestic use, as shown in the 2013 KEMA test, will not (including worst case scenario) exceed the RIVM life time exposure value of 0.05 µg/m^3.

The highest concentration measured during the KEMA sampling program was 6.9 µg/m^3 mercury in gas which would lead to an approximate peak exposure of 0.13 µg/m^3 in indoor air during 2 hours a day based on the scenario that the gas is burned in 3 periods of 20 minutes during the day.

Based on this model a poorly ventilated small house, which is supplied with gas containing a mercury concentration of 20 µg/m^3, would on a daily average still stay below the intervention value of 0.05 µg/m^3. The peak concentrations would, however, exceed this value.

RECOMMENDATIONS

This exposure model is based on the best information currently available. Future research regarding the default values for the variables used in this model should be carefully considered for the key variables: room size, mercury concentration in gas and ventilation. Whereas the last two mentioned variables could be subject to changes as a result of new or more elaborate findings.

In addition, one could test this theoretical model with indoor air quality experiment. Its aim would be to test whether the findings in this document relate to the actual indoor air qualities values one measures. Such an experiment requires expertise and thorough understanding of the technicalities of the design of the experiment and modeling conditions. Therefore, one could recommend that an authoritive organization, such as the RIVM, would be involved in such a test.

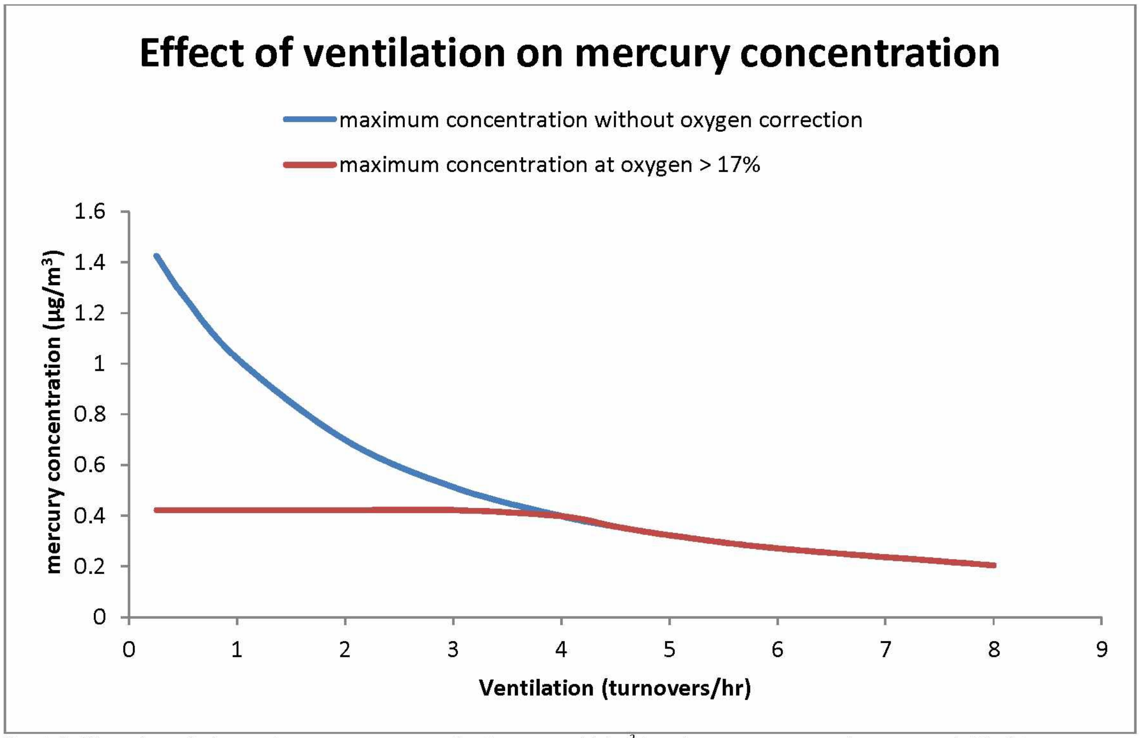
RIVM involvement is not required at this stage 1) Exposure model has been sufficiently cross checked with other research literature 2) Findings show that some level of mercury exposure can exist from Goningen gas in Dutch household, however, the average exposure level will not exceed the RIVM lifetime exposure limit 3) RIVM is a government organization which will publish its findings.

DOCUMENT VERSIONS

Version	Date	Author	Modifications
1	30-04-2014	5.1.2.e	Final version after peer reviews
2	14-01-2014	5.1.2.e / 5.1.2.e	Final version
1	2-08-2013	5.1.2.e	Generic assessment

APPENDIX I – Effect of ventilation on maximum mercury concentration in air for a room of 15m^3

The graph below shows the effect of ventilation on the maximum mercury concentration in air.
A room of 15m3 is used with a peak exposure of 1 hour.



Graph 2: Effect of ventilation on the mercury concentration in a room of 15 m^3 based on a gas consumption pattern of all in 1 hour.