Emissions from concrete – an indoor air quality issue?

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KEYWORDS: *Concrete, additives, emissions, ammonia, VOC, discoloration, indoor environment,*

SUMMARY:
Problems with indoor air quality, discolouration of oak flooring and odours are reported from newly constructed buildings. According to the industry emissions from concrete are more or less none existing. However on-site surveys in buildings with reported problems show that the concrete used in building constructions does emit volatile organic compounds, VOCs, and ammonia. Additives used in the concrete or unwanted impurities might be the source of the emissions.

Ammonia and VOC emissions levels have been measured on samples from buildings under constructions that were submitted to us for determination of RH. This study show that modern concrete, used in building construction today, does emit both ammonia and VOC even though it is presumably undamaged and does not have a surface layer coating. The majority of the samples have emission levels lower than 4 ppm for ammonia and 10 ppm for VOC. Diverging emission levels have mainly been found in samples from objects with a low water-cement ratio, which indicates that one or more of the ingredients in the concrete under certain circumstances can produce higher emission levels, however not all the samples from the same object with the same water-cement ratio have elevated emission levels hence more parameters than low water-cement ratio concrete seem to have an impact on the emission levels.

1. Introduction

Sweden historically has two well known emission damage types in concrete floor constructions. Both are initiated by elevated moisture levels due to insufficient drying during construction or later addition of moisture. One of the types is caused by degradation of casein (an amino acid) in self leveling compound that was common during a period (1974-84). The other well known floor damage type is the release of degradation products of plasticizers and other substances from glued flooring material with high moisture resistance. Indicator substances for the different damage types have been ammonia emissions from the concrete surface in the first case and 2-ethyl-1-hexanol and 1-butanol in the latter.

In recent years we have been asked to investigate the reason for discoloration of oak parquet floors, odour problems and/or health complaints in new dwellings on several occasions, in a few cases even during production. These objects neither had glued coating nor casein containing leveling compound. Nonetheless high levels of ammonia and volatile organic compounds (VOC) have sometimes been measured from the concrete, not only from the surface but also from samples taken deeper in the concrete slabs which indicate that the origin to the emissions this time is to be found within the concrete.

In recent years the use of additives has increased due to desired functionality, environmental considerations, time aspects in the building process and costs.

Our concern is that additives in concrete could be responsible for reported indoor environment problems possibly caused by unwanted synergetic reactions between different types of concrete

admixtures taking place in the reactive environment that concrete provides. Available information from the industry reports only low emissions from concrete.

The fact that the concrete formulas differ according to desired properties does not make the picture any clearer. There are references that modern concrete contains roughly 6-8 kg additives per m^3 (Naturvårdsverket, 1995, 2009), which means that the concrete in a normal sized room of 10 m^2, with a 200 mm thick concrete slag, contains 12-16 kg additives.

While the main ingredients in concrete (stone, gravel, sand, cement and water) are inorganic most concrete additives are organic and could therefore be a source of volatile organic compounds, in fact it has been shown that the emission from the concrete can occur when various additives are added to mixture (Johansson, 1994). The most commonly used additives are plasticizers that improve the workability of the concrete and thus also function as a water reducer. Additives often have unintended negative effect. According to Anderberg (2002) it was suggested by Byfors already in 1994 that preliminary investigations always should be made to ensure that unwanted effects do not occur. A Japanese study has shown that ammonia and VOC can be generated through alkaline hydrolysis within and emitted from hardened concrete (Tomoto, 2008). Urea-based antifreeze admixtures used in indoor concrete walls has been shown to be a source to indoor exposure to ammonia that may go on for a long period of time (Bai, 2006). Furthermore an unpleasant odor and mucous membrane symptoms have been linked to high levels of ammonia and benzene in a newly built office in Beijing (Lindgren, 2009)

Due to lack of information from the concrete producers regarding expected emission rates from the mixed concrete and the fact that samples submitted to us for analyses normally are from buildings with reported dampness and/or indoor air quality problems, our knowledge about what type of emissions and emission rates that can be expected from new undamaged concrete is insufficient. Therefore there is a need to establish normal emission levels (background levels) from concrete in newly constructed buildings without reported problems.

To get a better understanding of the type of emissions / emission levels expected from concrete in new buildings and also to compare some of the available, indicative methods normally used as a complement in on-site damage investigations, measurements have been performed on samples of concrete and floor leveling compound from new constructions.

2. Material and Methods

Measurements have mainly been performed on samples of concrete drill cores and in a few cases on floor leveling compound. All samples were taken from buildings during construction and, as earlier mentioned, were submitted to us for determination of moisture levels in terms of relative humidity (RH). None of the concrete samples have been subject to any kind of surface coating other than leveling compound and the leveling compound samples have not been coated at all.

In total we have performed measurements on 93 cores, 75 of concrete and 18 of floor leveling compound. RH and emission levels of ammonia, VOCs and water vapor have been measured on all the samples. The samples are from eleven different building projects with the number of samples included varying from one to 24.

2.1 Samples

As mentioned the samples were drilled out from buildings during production and submitted for RH-determination. The sampling have been carried out according to ‘RBK-metoden’, a standardized Swedish method for determination of RH in concrete structures, and the Swedish flooring industry's method for RH-determination in leveling compound respectively. The water-cement ratios for the different samples are known, with a few exceptions for the samples from hollow-core concrete

elements where w/c ratios of 0.4 or less have been assumed. No data regarding what additives that have been used have been available.

The concrete samples have, with a few exceptions, been taken from the equivalent measurement depth, which is defined based on the actual thickness and construction, while the leveling compound samples consist of the entire cross section. If, in the latter case, the sampled volume does not fit in a single test tube the material is mixed and divided into two test tubes. Hence in some cases there were two tubes with comparable content which allowed cross check evaluation of the used methods.

2.2 RH-determination

RH-determination has been carried out according to 'RBK-metoden' and the Swedish flooring industry's method for concrete and floor leveling compound samples respectively. The samples were collected prior to the application of the surface layer coating.

The uncertainty of these methods is approximately ± 2 % RH in the interval 75 – 95 % RH.

2.3 Ammonia, Organic compounds and Water Vapor in concrete, using modified OCIC method and B&K Multi Gas Monitor type 1302

The OCIC method is a modified head space technique where the sample is placed in a closed container which is kept at room temperature. The air in the pore system of the sample is allowed to reach equilibrium with the surrounding air which is then sampled and analyzed. The method was developed by Sjöberg (1998) to measure to what extent organic compounds, produced in the alkaline and moisture induced degradation process of plasticizers and other substances from carpet glue are deposited in concrete.

The sample is fractioned and placed in a gas tight 250 ml glass container fitted with stainless Swagelok tubing. The cover has an inner gasket of Teflon to prevent interference with the equilibrium process. This allows controlled extraction of air during sampling. The compounds in the sample and its pore system are allowed to reach equilibrium with the surrounding air in the glass container for 72-120 hours before air sampling.

Originally the sampling was carried out on a tube with Tenax TA sorbent which was analyzed for VOC with thermal desorption by gas chromatographic technique.

The method has been further modified and here the sampling was performed as follows: The sample was placed in the glass container after determination of RH, which in practice means minimum 4 days after it was drilled out, and left for 72-120 hours. The VOC and ammonia content in the equilibrated air were measured using the B&K Multi-gas monitor type 1302.

The B&K Multi-gas monitor type1302 is a gas analyzer. The principle of measurement is based on photo acoustic infra-red detection spectroscopy. The detection limit is gas-dependent but typically in the order of 10^{-3} ppm.

The VOC spectrum measured with B&K 1302 is broader than the normally used definition and includes VVOC, VOC and SVOC. Even though the filters used are calibrated for VOC (methane), ammonia and water vapor respectively cross sensitivity does occur. Measured levels are given with methane as reference. The sampling volume in each cycle is 140 cm^3.

The method in the applications used here should be viewed as an indicative method.

Before sampling an emptied glass tube is mounted directly on the Swagelok tubing outlet on the cover and connected to the instrument with a PTFE-tube. The inlet to the glass container is left open. The concentrations of ammonia, VOC and water vapor were measured. Before each measuring cycle, the background values were measured to ensure that one measuring cycle did not impact the next.

To evaluate the method the periods the samples were allowed to reach equilibrium before the first sampling have varied from a one up to 12 days. Where more than one cycle was measured new background levels were established before the next cycle.

FIG 1. Set up to measure VOC and ammonia with B&K 1302

Selected samples were left for an additional equilibration followed by measurement on Kitagawa detector tubes for ammonia.

2.4 Ammonia in concrete, using modified OCIC method and Kitagawa detection tubes

The preparations of the sample are the same as previously described. After the sample has been equilibrating air from the glass container is sampled on a Kitagawa detection tube for ammonia 0.2 – 20 ppm using the Kitagawa Gas aspirating pump AP-20.

As the detector tube does not have the right proportions to fit properly into the Swagelok coupling it has been fitted with thread tape and sealed with Parafilm to prevent leakage. When exposed to ammonia the color of the reagent changes from pink to whitish and the ammonia concentration in the sampled air is read on the scale of the tube.

The method in the applications used here should be viewed as an indicative method. The uncertainty in reading the detector tubes are reported to be ± 10-15%. Color change also occurs after exposure to other basic compounds such as organic amines, but with different scale.

FIG 2. Set-up to measure NH3 on detection tube

3. Results

A summary of the obtained results using the B&K 1302 modified methods for ammonia and OCIC and Kitagawa detection tubes on concrete samples can be found in table 1 and the distribution of values measured with B & K 1302 is shown in figure 3 below.

TABLE 1. Results from measurements using the B&K 1302 modified method for ammonia and OCIC and Kitagawa detection tubes on concrete samples.

	Sample age, days	NH_3, ppm B&K 1302	VOC, ppm B&K 1302	Water vapor, g/m³ B&K 1302	RH, %	NH_3, ppm Kitagawa
Minimum	11	0.476	3.77	11.9	61.2	2
Maximum	85	9.42	28.3	19.2	94.8	13
Mean	29.2	2.34	6.85	15.6	80.7	5.8
median	21	1.86	5.93	15.5	83.1	5
# samples		75	75	75	75	9

The majority of the measured concrete samples is comparable in both NH_3 and VOC respectively but diverges for some of the samples. 91 % of the sampled NH_3-values are below 4 ppm while 95 % of the sampled VOC-values are below 10 ppm.

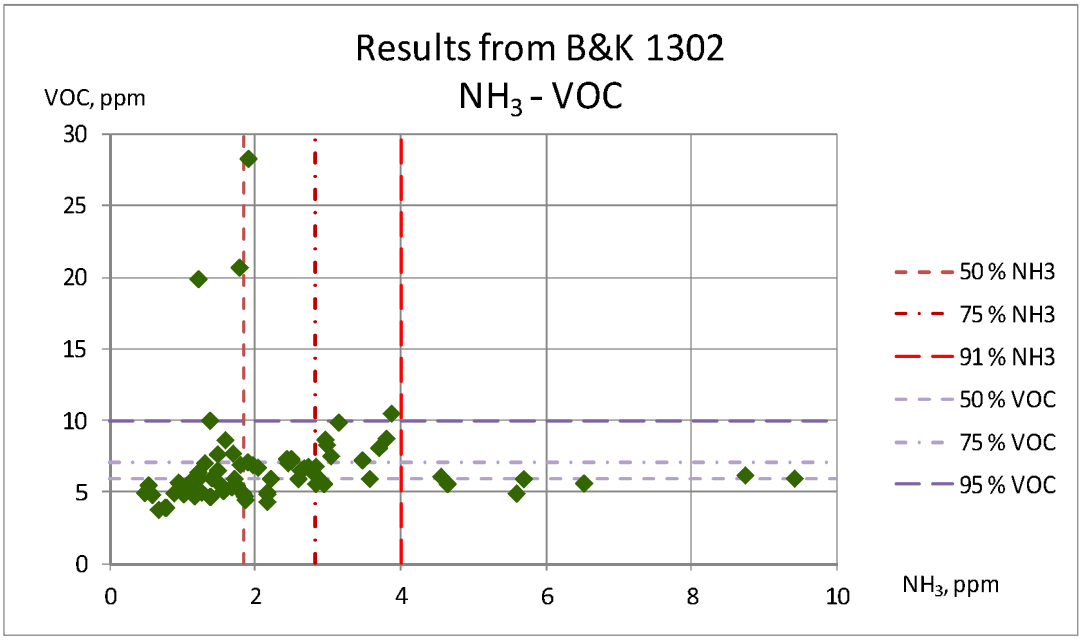

FIG 3. The distribution of values measured with B&K 1302 show that 50% are lower than 1.85 and 5.93 ppm, and 75% are lower than 2.83 and 7.1 ppm for NH_3 and VOC respectively.

The shares of measured values under 4 ppm for NH_3 and 10 ppm for VOC 95 and 97.5 % for samples with water-cement ratio higher than 0.4 and 85 and 95 % for samples with water-cement ratio 0.4 or lower respectively.

On nine of the samples ammonia emission levels were also measured on Kitagawa detection tubes. These measurements indicate that the correlation between the methods is relatively good, se figure 4.

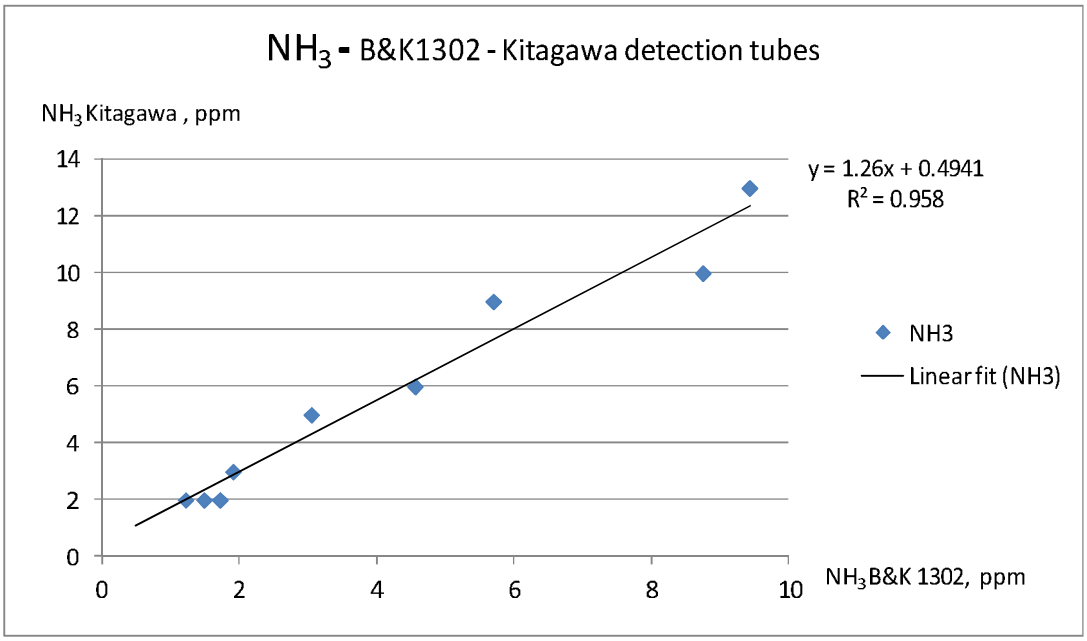

FIG 4. The correlation between NH_3values measured with B&K 1302 and Kitagawa tubes

The cross check evaluation of the double samples described in 2.1 shows that the modified method for OCIC with B&K 1302 is satisfyingly consistent for indicative measurements. Results are shown in table 2.

TABLE 2. Results from measurements using the B&K 1302 modified method for ammonia and OCIC and Kitagawa detection tubes on concrete samples.

Sample	NH_3, ppm A	NH_3, ppm B	Max/min ratio, NH_3	VOC, ppm A	VOC, ppm B	Max/min ratio, VOC	H_2O, g/m³ A	H_2O, g/m³ B	Max/min ratio, H_2O
1	1.94	2.43	1.25	9.65	10.70	1.11	19.80	19.10	1.04
2	2.41	1.98	1.22	11.40	9.61	1.19	19.10	17.40	1.10
3	2.23	2.40	1.08	7.21	6.07	1.19	17.60	16.70	1.05
4	2.21	2.00	1.11	9.94	7.36	1.35	17.00	17.00	1.00
5	3.22	3.02	1.07	18.40	17.00	1.08	19.00	19.40	1.02
6	3.11	3.45	1.11	11.70	11.60	1.01	19.50	20.30	1.04
7	1.09	1.23	1.13	5.81	5.33	1.09	12.40	12.70	1.02

The max/min ratio varies from 1.07-1.25, 1.01-1.35 and 1.00-1.10 for NH_3, VOC and water vapor respectively for the nine double samples with the corresponding mean values 1.14, 1.14 and 1.04.

4. Conclusions/discussion

In the present study, we have been referred to the submitted samples, one from each measuring point and each of them taken from an assigned depth in the structure. The recipe for the concrete regarding additives and all other parameters except water-cement ratios are unknown to us though we expect that they vary between, and in some cases maybe even within, the objects.

The results show that modern concrete, presumably undamaged, and with no surface layer coating, used in building construction today in fact does emit as well ammonia as VOC and that the emission levels in the majority of the so far investigated samples are less than 4 ppm ammonia and 10 ppm VOC. In this study this may be referred to as "normal concrete" in terms of emission levels. As the work proceeds and more samples are measured these reference values may have to be adjusted.

Considering that levels above 4-5 ppm NH_3 previously have been discussed as limits for discoloration of oak parquet flooring this is an interesting finding even though previous reference values were sampled with a different technique.

Among the results there are also a few samples with higher values for either ammonia or VOC which shows that high emission levels can arise from the concrete itself during the building process which indicates that one or more of the ingredients under certain circumstances can produce higher levels of emissions. An interesting fact is that while we have samples with higher emission levels of either ammonia or VOC, so far we have none with higher levels in both.

Some of the RH-values for the used samples are considerably lower than usually measured during the building process, 28 samples with RH lower than 80 %, however no disturbances were identified during the measuring process. As expected there is a correlation of water-cement ratio and RH but no connection could be seen between elevated emission levels and RH. Experiences from on-site surveys from buildings with reported discoloring of oak flooring and/or indoor problems as well as a small experiment with 150 mm concrete specimens indicate a correlation between high RH and high emission levels in samples from a specific cross section of the specimen. This study only includes 3 samples taken from the same cross section, these however indicates increasing values for both ammonia, VOC and RH which supports the earlier on-site survey findings and the theory that if other conditions are the same emission levels increases with moisture content.

Even though the number of samples with elevated emissions levels is small, especially for VOC emissions, a possible connection has been found regarding emission levels and water-cement ratio. In fact excluding the samples with water-cement ratio of 0.4 or lower also excludes most of the NH_3 values above 4 ppm and almost all the VOC values above 10 ppm. Yet another interesting fact is that while one of the objects, in all 24 samples with water-cement ratio 0.35 and obtained RH-values between 61.2 and 78.7 %, represents most of the samples with water-ratio of 0.4 or less and hence also most of the samples with elevated emission levels many of the other samples from this object had low emission levels of both ammonia and VOC.

While no specific information is available about the concrete formulas and additives used in these specific objects, a hypothesis is that low water-cement ratio concrete contains more additives, especially plasticizers, to increase the workability of the concrete and thus are more chemically complex. For instance plasticizers are based on organic compounds, concrete that contains more plasticizers potentially could emit more VOC. The fact that one object of 24 samples includes four samples with elevated levels of ammonia, three with VOC and the other 17 with low emissions of both indicates that while the chemical content, the additives or impurities in the concrete, appears to be a main contributor there are also other parameters that have a major impact on the outcome concerning emission levels.

Regarding VOC we still do not know enough to connect obtained values to potential indoor environment problems. The results from this study will give a better understanding of the expected emission levels of ammonia and VOC from new undamaged concrete and possibly what emission levels that are to be considered elevated and thus potentially could pose a negative impact on the indoor environment.

In addition to the results that are presented here, our intention is to carry out on-site FLEC measurements on concrete as well as OCIC on Tenax TA to link these to our modified methods of measurement and also to identify what compounds the emissions from new concrete contains.

The modified method for OCIC-measurements with B&K 1302 has been shown to work well for indicative measurements. The values from the floor leveling compound samples show good compliance for the double samples. Further analysis needs to be done to specify the method concerning the time period needed to optimize the equilibrating and to minimize leakage. The

comparison between measuring ammonia levels with B&K 1302 and Kitagawa detection tubes show good correlation for concrete samples, with a tendency for higher values for the detection tubes.

Questions of concern, among others, for the future are what emissions can be expected from concrete, in what way the use of fly ash in the concrete will affect the emissions and emission rates, what other chemicals will be added to the concrete? We believe there is a need that the concrete producers provide adequate information concerning expected emission levels for their concrete formulas including additives.

5. Acknowledgements

This work was carried out with support from our employer AK-konsult Indoor Air AB. The support, encouragement and understanding from our employer and our colleagues are gratefully acknowledged.

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