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Research article



## MATHEMATICAL MODELING OF AMMONIA EMISSION RATE IN NEWLY CONSTRUCTED BUILDINGS

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A rapid growth in monolithic residential construction over recent decades has created a problem associated with ammonia contamination inside newly constructed buildings. Absence of substantiated preventive actions aimed at minimizing ammonia emissions hinders commissioning of new residential buildings and may create an unfavorable sanitaryepidemiological situation with obvious olfactory-reflex and irritating effects on public health.

The aim of this study was to develop a scientifically grounded method to predict when ammonia concentrations emitted from concrete constructions would reach their permissible levels in air inside contaminated premises in newly constructed buildings.

Ammonia emissions were estimated based on data of laboratory tests that involved analyzing indoor air samples taken in Saint Petersburg and the Leningrad region. Indoor air was analyzed in 4 newly constructed residential buildings (165 premises, 57 test protocols, 893 air samples tested to identify ammonia in them). Relationships between changes in ammonia concentrations and ventilation time were obtained by using regression analysis (regression equation, least square method). To establish reproducibility of the results and a possibility to compare them, we tested variances for homogeneity by using Fisher criterion. Sampled populations were compared with Student's t-test in case the data fitted to a normal distribution (Kolmogorov – Smirnov test, Shapiro – Wilks test). Critical significance was taken at 0.05 in all the statistical comparisons.

We have developed a method for predicting when ammonia concentrations that occurred in indoor air inside newly constructed buildings due to multi-day emissions from building materials would reach their permissible levels. The method involves multi-day measurements (y,  $mg/m^3$ ) of ammonia concentrations sequentially in each premise inside a newly constructed building on any day of measurements during the time period t; building up relationships between averaged ammonia concentrations ( $y_{av}$ ,  $mg/m^3$ ) and a time moment t; mathematical analysis of the obtained relationships by parameterization and statistical analysis of the obtained kinetic parameters.

**Keywords:** monolithic residential buildings, indoor air, ammonia, ammonia emission, mathematical modeling, building materials, concrete.

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A rapid growth in residential construction based on using monolithic reinforced concrete as a building material has created a problem associated with ammonia contamination in indoor air. This contamination is largely caused by ammonia compounds that are contained in raw components of concrete mixes. These compounds are introduced with chemical modifiers that are usually added to concrete and concrete mixes (hardeners, antifreeze admixtures etc.) or with auxiliary components in concrete production (grinding intensifiers). Industrial wastes used as mineral fillers (slag or ash) can be another source of ammonia in indoor air [1–12].

Ammonia is a result of hydrolysis and is generated from amides, amines and ammonia compounds.

Studies that addressed ammonia emission from concrete into indoor air have been accomplished in many countries; their results indicate the chemical occurs in indoor air in considerably high concentrations. Thus, Z. Bai with colleagues (2006) established ammonia concentrations varying between 2.30 and 5.85 mg/m<sup>3</sup> in indoor air inside newly constructed buildings; these concentrations occurred due to use of urea-based antifreeze admixtures in concrete [8]. T. Lindgren (2010) described a case when an elevated ammonia concentration was detected in a newly built office in Beijing and this was also due to antifreeze admixtures in concrete blocks [10].

Russian studies report similar results. D. Fokin (2011) showed in his work that ammonia migration from walls in a newly built residential building made of monolithic reinforced concrete created indoor air contamination at a level equal to  $2 \text{ mg/m}^3$  (50 MPCav.d.). The author pointed out that ammonia concentrations in indoor air were not influenced by furniture or decoration [13].

The work [4] describes the results obtained by instrumental measurements of indoor air inside residential and public buildings that were accomplished in Saint Petersburg and the Leningrad region over the period between 2012 and 2018. These tests were both scheduled or performed as per developers' and / or citizens' claims. Ammonia was detected in concentrations higher than MPCav.d. in 1147 samples out of 2839 (40.4 %). The established levels were by 163 times higher than the existing hygienic standards (MPCav.d.) in some cases. Ammonia was fixed in a concentration lower than sensitivity of the applied chemicalanalytic method only in 1.1 % of the analyzed samples.

Intensity of ammonia migration from concrete into indoor air, just as any other chemical, depends on air temperature and humidity and on volumes of construction materials present in a given premise [1, 3]. Z. Bai with colleagues (2006) showed in their studies that an increase in air temperature leads to more intense ammonia emission and to higher ammonia emission rates. Besides, an ammonia concentration directly depends on ventilation in a premise. According to the work [8], it takes more than ten years to reach complete ammonia emission from concrete that contains urea-based admixtures. However, there have been no studies so far with their focus on determining a period over which ammonia concentrations in indoor air drop down to safe levels regulated by hygienic standards.

Ammonia in air has been established to produce olfactory-reflex and irritating effects even in small and average concentrations. High concentrations can induce acute poisoning. An issue related to providing safe environment in residential buildings given ammonia emissions from building materials is quite urgent; this is confirmed by multiple complaints about indoor air in residential and public buildings made by citizens and workers to federal and regional executive authorities [4, 7–19].

Although the issue is truly vital, there are still no available scientifically substantiated methods to predict how long it will take ammonia concentrations emitted from concrete construction in indoor air to fall down to their permissible levels. This hinders commissioning of new residential buildings [4, 12–14, 16, 20–24].

In this study, our aim was to develop a scientifically grounded method to predict when ammonia concentrations emitted from

concrete constructions would reach their permissible levels in air inside contaminated premises in newly constructed buildings made of monolithic reinforced concrete.

**Materials and methods.** Relationships between ammonia concentrations in indoor air and time were built and analyzed by using 57 test protocols that described the results of laboratory tests of indoor air in buildings in Saint Petersburg and the Leningrad region. The tests were performed by the Hygiene and Epidemiology Center of Saint-Petersburg and Leningrad region.

Air tests were performed following citizens' complaints about unpleasant smells as well as within production control of buildings that were to be commissioned. Overall, 893 air samples were taken in 165 premises to identify ammonia in them and 285 tests were performed.

Sampling was accomplished with electrical aspirators OP-824 TZ in accordance with the State Standard GOST R 57256-2016<sup>1</sup>. The process involved three sequential measurements of ammonia concentrations in air 8 hours after 15 minutes of cross ventilation with fully opened windows done once a day for 20 minutes.

Ammonia mass concentrations were measured as per a procedure based on "catching ammonia in air with an acid solution and its photometric detection as per indophenol..."<sup>2</sup>.

The research objects were four apartment houses (hereinafter called objects) with their

load-bearing walls made of monolithic concrete. All four objects had natural ventilation that provided intense airing inside them. Air was tested to identify ammonia in it in all four objects. The tests in objects No. 1 and 2 were performed prior to their commissioning; in objects No. 3 and 4, after the commissioning. The objects were at different stages regarding completion of decoration works inside them.

There was no interior decoration inside the objects No. 1 and 2. Floors and ceilings were made of concrete slabs; walls were made of concrete and foam concrete blocks. Premises inside the objects No. 3 and 4 had finished interior decoration and 6 premises already had some furniture.

Air sampling was made considering preliminary identification of microclimate indicators that conformed to the established sanitaryepidemiological requirements. It was done to eliminate any influences exerted by environmental factors that would facilitate ammonia emission from concrete.

Mathematical analysis and parameterization were accomplished by using the least square method (non-linear regression) as per the equation (1); Statistica 10 software package was used in the process<sup>3</sup>. That is, we determined average values of the basic kinetic parameters  $A_{av}$ ,  $B_{av}$  and  $C_{av}$  for all the relationships established for all the objects:

$$y_{av}(t) = A_{av} \cdot \exp\left(-B_{av} \cdot t\right) + C_{av}, \qquad (1)$$

<sup>&</sup>lt;sup>1</sup> GOST P 57256-2016. Indoor air. Sampling strategy for ammonia (approved and validated by the Order of the Federal Technical Regulation and Metrology Agency issued on November 10, 2016 No. 1664-st). *KonsultantPlus: electronic fund for legal and regulatory documents*. Available at: https://docs.cntd.ru/document/1200141431 (May 22, 2022) (in Russian).

<sup>&</sup>lt;sup>2</sup> RD 52.04.186-89. Rukovodstvo po kontrolyu zagryazneniya atmosfery (utv. Zamestitelem predsedatelya Goskomgidrometa SSSR 01.06.1989, Glavnym gosudarstvennym sanitarnym vrachom SSSR 16.05.1989); red. ot 11.02.2016, s izm. ot 03.07.2020 [Guide 52.04.186-89. Guide on control of ambient air pollution (approved by the deputy to the Head of the USSR Gosgidrokomitet on June 01, 1989, the USSR Chief Sanitary Inspector on May 16, 1989); last edited on February 11, 2016, last amended on July 03, 2020]. *KonsultantPlus: electronic fund for legal and regulatory documents*. Available at: https:// docs.cntd.ru/document/1200036406 (May 22, 2022) (in Russian).

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where  $y_{av}(t)$  is the exponential relationship between a decline in ammonia concentration  $y_{av}$ in indoor air in residential premises and time *t*;

 $A_{av}$  is the average value of the total change in ammonia concentration  $y_{av}$  over the whole analyzed time t, mg/m<sup>3</sup>, the parameter identified by the graph analysis;

 $B_{av}$  is the average value of the decline rate constant for ammonia concentration  $y_{av}$  in indoor air in residential premises, the parameter identified by the graph analysis and associated with the essence of the migration process, ammonia emission from concrete materials and a material itself, day<sup>-1</sup>;

 $C_{av}$  is the average value of the minimal residual ammonia concentration in indoor air at the end of the experimental period, mg/m<sup>3</sup>, the parameter identified by the graph analysis.

The formula (1) establishes the exponential relationship between a decline in ammonia concentration  $y_{av}$  in indoor air in residential premises and a time *t*; that is, it describes ammonia emission from building materials over time. The equation (1) with average values of the basic parameters  $A_{av}$ ,  $B_{av}$  and  $C_{av}$  calculated with the least square method describes a parameterized approximated curve. This curve goes through all the experimental points in the graph in the best way.

We calculated expanded uncertainty (inaccuracy) U(T) of the value T (in days) for a given object as per the formula (2) at p < 0.05:

$$U(T) = \frac{\sigma_T}{\sqrt{3 \cdot n}} \cdot t_{P=0.95; f=n-1}, \qquad (2)$$

where the standard deviation  $\sigma_T$  was calculated by summing all the standard uncertainties considering three contributions made by the standard deviations  $\sigma_A$ ,  $\sigma_B$  and  $\sigma_C$  of all three kinetic parameters  $A_{av}$ ,  $B_{av}$  and  $C_{av}$ , calculated within the analysis, the very value *T* depending on them:

$$\sigma_T = \frac{1}{B_{av}}.$$
 (3)

$$\cdot \sqrt{\left(\frac{\sigma_B}{B_{av}}\right)^2 \cdot \left(\ln \left|\frac{C^* - C_{av}}{A_{av}}\right|\right)^2 + \left(\frac{\sigma_{Cav}}{C^* - C_{av}}\right)^2 + \left(\frac{\sigma_A}{A_{av}}\right)^2},$$

where  $C^*$  is the selected standard concentration, for example, equal to MPCav.d.; the declining extrapolated curve crosses with it at the time moment T.

**Results and discussion.** Average initial ammonia concentrations  $C_0$  (mg/m<sup>3</sup>) in indoor air were considerably higher than its MPCav.d. in premises inside all the analyzed objects; that is,  $C_0$  / MPCav.d. ratio was within a range between 3.55 to 30.4 in different premises.

Ammonia concentrations were measured in indoor air in all four objects within a multiday study; as a result, we observed a monotonically decreasing relationship between an ammonia concentration and time, which is shown as a descending curve on the graph.

The Figure provides an example of a typical curve describing a relationship between an ammonia concentration  $y_{av}$  in indoor air and a time *t* for the object No. 1 (each point represent an average value out of three or six ammonia concentrations *y* in air inside each premise).



Figure. A relationship between an ammonia concentration in indoor air in residential premises inside the object No. 1 and time (all the measures were performed under identical conditions)

For the object No. 1, as well as for all the other analyzed objects, the created graphic relationships between average values  $y_{av}$  of ammonia concentrations (in mg/m<sup>3</sup>) and an emission time *t* are exponential curves that asymptotically tend to the average minimal concentration  $C_{av}$ . All the relationships satisfy the equation (1). Therefore, the analysis made it possible to take the exponential relationship between a decline in an ammonia concentration  $y_{av}$  in

## Table

The analyzed object	Average initial concentration, $(A+C)^*$ , mg/m <sup>3</sup>	Average initial concentration to MPCav.d. ratio	Total change in concentration $A^*$ , mg/m <sup>3</sup>	Observed change rate constant $B^*$ , days <sup>-1</sup>	Residual concentration $C_{av} \pm \Delta C^*$ , mg/m <sup>3</sup>	A time moment when MPCav.d. was reached <i>T</i> *, days
1	$0.144\pm0.023$	3.6	$0.138\pm0.02$	$0.02209 \pm 0.00742$	$0.0065 \pm 0.0121$	$64 \pm 11.2$
2	$0.145\pm0.01$	3.55	$0.122\pm0.008$	$0.02419 \pm 0.00483$	$0.0228 \pm 0.00513$	$81.2\pm8.7$
3	$0.163\pm0.01$	3.98	$0.145\pm0.005$	$0.01947 \pm 0.00891$	$0.01784 \pm 0.0084$	$96.4\pm28.6$
4	$1.24 \pm 0.141$	30.4	$1.215\pm0.134$	$0.01736 \pm 0.00463$	$0.02475 \pm 0.0451$	$252.2 \pm 88.3$

Time moments when ammonia concentrations reached their permissible levels at the objects No. 1–4

indoor air inside premises and a time t as a physical law that describes ammonia emission from building materials and is mathematically given with the equation (1). In future this law can be applied to analyze and parameterize all the experimentally obtained points in graphic relationships.

The Table above provides all the calculated average values of the basic kinetic parameters  $A_{av}$ ,  $B_{av}$  and  $C_{av}$  for all the relationships established for all the analyzed objects and their uncertainties (inaccuracies).

As an ammonia concentration  $y_{av}$  in indoor air changes, at a certain time moment t, which is equal to T, it can reach any target permissible ammonia concentration  $C^*$  in indoor air, including the established maximum permissible level (MPL)  $C^*$  for ammonia equal to 0.04 mg/m<sup>3</sup>. We can assume a time moment T when the kinetic curves reach any permissible ammonia concentration  $C^*$  relying on values of the kinetic parameters  $A_{av}$ ,  $B_{av}$  and  $C_{av}$  in the equation (1). In particular, this level can be equal to MPC established for ammonia, which is 0.04 mg/m<sup>3</sup>.

We determined a time moment T when the kinetic curves reach MPC for ammonia equal to 0.04 mg/m<sup>3</sup> for each object as per the equation (4) using numerical values of the parameters  $A_{av}$ ,  $B_{av}$  and  $C_{av}$  in the equation (1) of the experimental relationships established for each analyzed object:

$$T = -\frac{1}{B_{av}} \cdot \ln \left| \frac{C^* - C_{av}}{A_{av}} \right|, \tag{4}$$

where T is a predicted time moment when a permissible concentration  $C^*$  of ammonia

emitted from building materials into indoor air inside a new building is reached, starting from the first day of measuring ammonia concentrations, days;

 $B_{av}$  is the average value of the decline rate constant for ammonia concentration  $y_{av}$  in indoor air in residential premises, the parameter identified by the graph analysis and associated with the essence of the migration process, ammonia emission form concrete materials and the material itself, days<sup>-1</sup>;

 $C^*$  is the established permissible ammonia concentration, mg/m<sup>3</sup>, for example, MPC;

 $C_{av}$  is the average value of the minimal residual ammonia concentration in indoor air at the end of the experimental period, mg/m<sup>3</sup>, the parameter identified by the graph analysis.

 $A_{av}$  is the average value of the total change in ammonia concentration  $y_{av}$  over the whole analyzed time t, mg/m<sup>3</sup>, the parameter identified by the graph analysis.

We determined the time T for each object as per the equation (4) as follows:

1) The object No. 1:

$$T = -\frac{1}{0.02209} \ln \left| \frac{0.04 - 0.00653}{0.13759} \right| = 64 \text{ days};$$

2) The object No. 2:

$$T = -\frac{1}{0.02419} \ln \left| \frac{0.04 - 0.02281}{0.12251} \right| = 81.2 \text{ days};$$

$$T = -\frac{1}{0.01947} \ln \left| \frac{0.04 - 0.01784}{0.14479} \right| = 96.4 \text{ days};$$

4) The object No. 4:

$$T = -\frac{1}{0.01736} \ln \left| \frac{0.04 - 0.02475}{1.215} \right| = 252.2 \text{ days.}$$

The values T calculated with the equation (4) and their inaccuracies calculated as per the formula (2) for all the analyzed objects are given in the Table.

We established that in case the initial conditions are the same (average initial ammonia concentrations in indoor air are almost identical and do not exceed 4 MPCav.d.) a time period up to 125 days (according to the upper confidence limit) ensures that an ammonia concentration falls below MPCav.d. in premises without interior decoration (the objects No. 1 and 2) as well as in variable premises with decoration and furniture (the object No. 3).

High values of the time period T that is necessary for ammonia concentrations to reach MPCav.d (8–11 months) at the object No. 4 are due to the initial ammonia concentrations (A + C) that were substantially higher than at three other objects (more than 30 MPCav.d.).

Following the research, the invention No. 2760762 was patented in the RF as "The method for predicting a time moment when concentrations of ammonia that occur in indoor air inside newly constructed buildings due to multi-day emissions from emitted from building materials will reach their permissible level"<sup>4</sup>.

**Conclusions.** Therefore, we established that an exponential equation should be used

to describe ammonia emission from building materials into indoor air inside newly constructed buildings associated with time of ventilation. The suggested method for predicting how ammonia concentrations will decline in new premises has been developed using mathematical procedures and provided with statistical substantiation. This makes it possible to use it to analyze any experimentally established relationship to estimate ammonia emission from building materials into indoor air.

The identified quantitative kinetic parameters of the equation for approximated curves give an opportunity to calculate ammonia emission rate and a time period over which ammonia concentrations will fall down to their permissible levels (MPC).

The suggested method for predicting a time moment when ammonia concentrations fall down to their permissible levels makes it possible to effectively control safety of indoor air and to establish when residential and public buildings should be commissioned provided that they do not pose any health harms for people.

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