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The Drawback of Optimizing Air Cleaner Filters for the Adsorption of Formaldehyde

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Abstract: Air cleaners with activated carbon (AC) filters for the adsorption of gaseous pollutants are often used to improve indoor air quality. As formaldehyde is a common and health-relevant indoor air pollutant, many testing standards for air cleaners, such as GB/T 18801:2015, require the cleaning efficacy to be tested with this substance. This often persuades manufacturers to optimize the employed filters specifically for formaldehyde. However, in regions where indoor formaldehyde levels are far below the guideline values, other gaseous pollutants might be more relevant. Thus, the question arises of whether the optimization for formaldehyde can have a negative impact on the adsorption of other gases. To address this question, the clean air delivery rate (CADR) of an air cleaner was determined for different test gases with either a standard AC filter or an AC filter modified for improved formaldehyde adsorption. Although the modified AC filter performed substantially better for formaldehyde, a strong reduction in the CADR was observed for toluene and nitrogen dioxide. This is a drawback for situations in which these gases are more problematic than formaldehyde. The findings suggest using either specialized filters for different applications or blends of different adsorbants to find the best compromise for the most relevant pollutants.

Keywords: air cleaner; adsorption; activated carbon; formaldehyde; air quality

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1. Introduction

Exposure to gaseous air pollutants is known to adversely affect human health [1,2]. Among the large spectrum of relevant gases, special attention is paid to volatile organic compounds (VOCs), which are common contributors to poor indoor air quality, as they can originate from various indoor and outdoor sources. As modern buildings become increasingly airtight to improve energy efficiency, high VOC concentrations are expected to become more common in residences if no additional measures are applied [3]. Among the VOCs, formaldehyde (IUPAC nomenclature methanal, HCHO) is one of the most studied compounds because of its ubiquity and well-established human health significance [4]. Formaldehyde is classified as a group 1 carcinogen according to the World Health Organization (WHO) International Agency for Research on Cancer (IARC) [5]. Furthermore, it can cause irritation of the eyes, upper airways, and skin [6]. Prolonged formaldehyde exposure has been associated with reduced pulmonary function [7] and asthma [8]. Primary formaldehyde emitters are paints, wood-based materials such as laminate and furniture [9], textiles, and consumer products [10]. Moreover, formaldehyde can result from smoking or incomplete combustion associated with gas stoves or wood-burning fireplaces [11]. Besides residential homes, workplaces can also be a relevant environment for exposure to formaldehyde [12,13]. For US residences, formaldehyde has been estimated to have the fourth highest chronic health impact of air pollutants after particulate matter with an aerodynamic diameter of <2.5 µm ($PM_{2.5}$), second-hand smoke, and radon [14]. Globally, among the gaseous pollutants, formaldehyde causes the highest loss of disability-adjusted life years (DALYs), which is a common metric to quantify and rank the burden of household air pollution [15].

Consequently, the WHO has established an indoor quality guideline for the formaldehyde concentration of 80 ppb (0.1 mg m^{-3} at 1013.25 hPa and 25 °C), which applies to all 30 min periods lifelong [16]. If this recommendation cannot be met, there is the option to either mitigate the sources or reduce the airborne concentration by diluting with clean air or by applying gas-cleaning technologies. Where natural ventilation is not possible and no ventilation systems are installed, air cleaners offer an alternative. These mobile devices have a fan that draws in the room air, passes it through one or more filters, and releases the cleaned air back into the room [17]. Air cleaners have become increasingly popular in recent years, as they are known to reduce the concentration of infectious aerosols [18–21]. However, many air cleaners are also equipped with activated carbon (AC) filters to reduce the concentration of gaseous pollutants [22–24].

Adsorption by AC is a common technology for removing gaseous pollutants because it is easy to operate and relatively inexpensive. However, although AC exhibits a high adsorption capacity for most gaseous pollutants, it cannot efficiently adsorb formaldehyde [25]. One reason is that the low boiling point of formaldehyde of 254 K hinders pore condensation even within highly porous adsorbents [26]. Moreover, the polarity of formaldehyde reduces the chemical interaction with the non-polar AC [25]. To improve the adsorption efficiency, surface modifications of the AC are often employed. Especially, the introduction of nitrogen groups was found to increase the formaldehyde adsorption efficiency [27,28]. The addition of nitrogen-containing functional groups can. for example, be efficiently achieved by impregnation with amines [29–32]. A different approach to increase the overall adsorption efficiency is to blend the AC in the filter with other adsorbants, which have previously been shown to be efficient for formaldehyde, such as zeolites, mesoporous silica, or metal–organic frameworks [33,34].

Although various studies have shown the positive effect of such modifications on formaldehyde adsorption, the extent to which this optimization might negatively affect the adsorption of other gases has not yet been investigated. If there were such detrimental effects, this would be an important finding since air cleaners are expected not to adsorb only a single gas but a variety of gaseous pollutants. For example, a study in more than 600 German households showed that the formaldehyde limit values were exceeded in only 0.1% of the cases [35]. Furthermore, formaldehyde concentrations have shown a decreasing trend over the years in European and Chinese residences [3,36]. By contrast, other VOCs are still present at high concentrations [37].

Another typical indoor gaseous air pollutant is toluene (IUPAC nomenclature: methylbenzene, C_7H_8). Toluene is one of the most abundant VOCs in indoor environments [38,39], as it is widely used in industry and in a number of commercial products, such as cosmetics, inks, adhesives, paints, and glues [40]. Therefore, and because it is a less hazardous model compound for benzene, toluene is prescribed as a test substance in testing standards for adsorptive indoor air purification, such as ISO 10121-2:2013 [41]. It is a well-known neurotoxicant [42] and mainly affects the central nervous system, causing fatigue, headache, eye irritation, or memory impairment [43]. A recent study reviewing different international occupational exposure limit values recommended an 8 h time-weighted average value of 20 ppm and a 15 min short-term exposure limit of 100 ppm [44].

Besides VOCs, inorganic gases, such as nitrogen dioxide (NO_2), can also have a high relevance for indoor air quality. NO_2 can either infiltrate from the outdoor air, where it is mainly generated from fossil fuel combustion, or result from indoor combustion processes, such as gas cooking, candles, or smoking [45]. A review of cohort studies found positive associations between long-term concentrations of NO_2 and mortality [46]. Furthermore, epidemiological studies have revealed that exposure to NO_2 in early life may lead to allergic diseases, including asthma, and have long-term effects on lung function [47]. Consequently, in 2021, the WHO lowered its guideline value for the annual mean concentration to 10 μg m^{-3} [48]. Because of its importance as a pollutant, NO_2 is also specified as a test gas in ISO 10121-2:2013 [41].

To reveal in an exemplary case whether the optimization for formaldehyde can be a drawback for other gases, this study compared two different filters in the same air cleaner, one with pristine AC and one with the same AC filter modified for improved adsorption of formaldehyde. The cleaning performances for formaldehyde, toluene, and NO_2 were compared to draw conclusions on possible side effects of the optimization.

2. Materials and Methods

As a measure of the cleaning efficacy, the clean air delivery rate (CADR) defined in GB/T 18801:2015 [49] and other standards, such as ANSI/AHAM AC-1:2020 [50] and IEC 63086-1:2020 [51], was used. The CADR indicates the volume of cleaned air provided by the air per time unit and ideally corresponds to the product of filter efficiency and flow rate. To determine the CADR, a test gas is introduced to a sealed test chamber until a certain starting concentration is reached. After homogeneous mixing is achieved, the exponential decrease in concentration:

$$C(t) = C_0 e^{-kt} \quad (1)$$

is measured over time with the air cleaner running. A decay curve of the gas concentration without the air cleaner serves as a reference to separate the natural decay from that caused by the air cleaner. The CADR is calculated as the product of the difference between the two decay rates with and without the air cleaner $(k_{tot} - k_{nat})$ and the volume V of the test chamber:

$$\mathrm{CADR} = (k_{tot} - k_{nat}) \cdot V \quad (2)$$

The CADR measurements were performed in a test chamber according to GB/T 18801:2015 [49], which is schematically shown in Figure 1. The internal dimensions of the chamber were 3.45 m × 3.40 m × 2.50 m, corresponding to a volume of 29.3 m³. To reduce the concentration of gaseous pollutants prior to the test, an air cleaner with a known high efficiency for all test pollutants was initially operated. Prior to each test, the temperature was set to (25 ± 3) °C with a wall-mounted air conditioner, and the relative humidity was set to (50 ± 10)%rh with a portable humidifier. These values, defined in GB/T 18801:2015 [49], can be regarded as representative of a typical indoor situation. However, we note that the following findings might slightly depend on the chosen relative humidity. For example, it is known that the adsorption capacity for toluene is reduced at a higher relative humidity [52], while the effect is less pronounced for NO_2 [53]. For formaldehyde, either positive or negative effects of increased relative humidity on the adsorption capacity were found depending on the details of the AC [26,54]. These complex dependencies can lead to relative shifts between the adsorption efficiencies of different adsorbates. However, it was beyond the scope of this work to investigate the effects as a function of relative humidity.

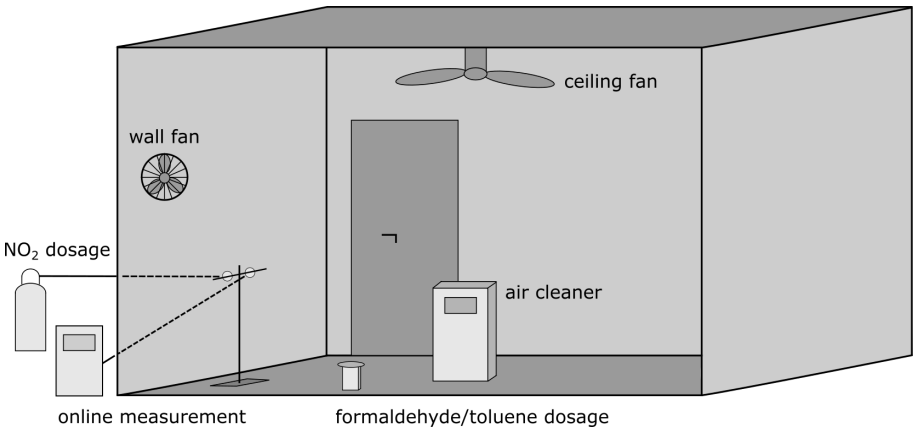

Figure 1. Schematic of the CADR test chamber.

During the actual measurements, the additional air cleaner, the air conditioner, and the humidifier were turned off. A wall fan at a height of 1.50 m and a distance of 0.40 m from the wall was operated during the whole test to mix the air in the chamber. While feeding the gas to the chamber, a centrally-mounted ceiling fan was also operated. The use of the mixing and ceiling fan allowed for reproducible testing conditions. However, it was previously demonstrated that such standardized tests are also representative of most typical real indoor environments without active ventilation [55,56]. The expanded uncertainty of the measurement method was estimated at ±10% in accordance with ISO/IEC 17025:2017 [57]. Sampling was performed by extracting a small flow from the chamber to the measuring instruments. The sampling point was located 0.50 m from the wall and 1.20 m above the floor.

The device under testing was a commercial air cleaner (model Philips AC 2939/10, Amsterdam, The Netherlands). It contained a cylindrical filter (model Philips FY 2122, Amsterdam, The Netherlands) consisting of a pleated fibrous filter medium for particle filtration with integrated AC grains for gas adsorption. According to the datasheet, the nominal CADR of formaldehyde is 220 m^3/h. Besides the original filter, which contains an additive to optimize the performance for formaldehyde, the manufacturer provided a derived sample filter that did not contain the chemical modification. For confidentiality reasons, it is not known to the authors what chemical modification was applied to the filter to improve the formaldehyde adsorption. In the following, the two filter types are denoted as "pristine" and "modified" filters.

To exclude effects from adsorption or desorption of water during the tests, all filters were first dried in a climate cabinet at 60 °C for 24 h and conditioned afterward for 24 h at 25 °C and 50%rh. For the tests, the air cleaner was placed in the center of the test chamber on the floor and at the highest continuous operation mode (level 3) at a constant input voltage of 230 V using a stabilized power supply.

For the three test gases, the following methods for dosing and measuring the concentration were applied. The initial concentrations were chosen according to GB/T 18801:2015 [49], which defines them as (10 ± 2)-times the Chinese limit values for indoor air specified in GB/T 18883:2002 [58].

- The initial formaldehyde concentration of about 815 ppb (1.0 mg m^{-3} at 1013.25 hPa and 25 °C) was generated by the sublimation of paraformaldehyde (>95% purity) at 200 °C. The concentration was measured with a continuous formaldehyde monitor (model AeroLaser AL 4021). The detection was based on the fluorescence of the product formed in the reaction of gaseous formaldehyde trapped in an aqueous solution with different liquids (Hantzsch reaction).
- The initial toluene concentration of about 530 ppb (2.0 mg m^{-3} at 1013.25 hPa and 25 °C) was generated by the evaporation of liquid toluene (99.8 % purity) on a glass tray at an ambient temperature of 25 °C. The concentration was measured with a compact proton-transfer-reaction mass spectrometer with a quadrupole mass analyzer (PTR-MS, model IONICON Compact PTR-MS).
- The initial NO_2 concentration of about 1280 ppb (2.4 mg m^{-3} at 1013.25 hPa and 25 °C) was generated by feeding NO_2 from a gas cylinder (10,000 ppm NO_2 in nitrogen 5.0) into the test chamber. NO_2 was measured with a chemiluminescence detector (CLD, model Environnement AC 32M). Besides NO_2, the CLD detector also measures the concentration of nitrogen monoxide (NO).

After the initial concentration was reached, the air was mixed for another 10 min to achieve homogeneous mixing. After that, either the natural decay was recorded for 60 min or the air cleaner was switched on to measure the total decay for 60 min. The data were exponentially fitted according to Equation (1) to derive the decay rates and CADR.

In addition to the CADR tests with gases, CADR tests with cigarette smoke particles in the size range of 0.3 µm to 10 µm were also performed in accordance with GB/T 18801:2015 [49] to estimate the flow rate of the air cleaner. The details of the test method are described in Ref. [17].

3. Results

3.1. Reference Measurements with Cigarette Smoke Particles

Taking into account that the filter has a particle filtration efficiency close to 100% (99.97% for 0.003 µm NaCl particles, according to the datasheet), the CADR for particles can serve as an estimate for the flow rate of the appliance. For the pristine filter, a CADR of 341 m^3h^{-1} was determined with cigarette smoke particles, while the modified filter revealed a CADR of 355 m^3h^{-1}. As the difference was within the uncertainty of the test method, it can be assumed that the modification did not cause significant macroscopic changes to the filter material, which would alter the pressure drop and thus also the CADR via the flow rate. Therefore, it is a reasonable assumption that the following tests with gases were performed at approximately the same flow rate for both filters.

3.2. Measurements with Formaldehyde

Figure 2 shows the decay curves measured with formaldehyde for the two filter types. The top and bottom rows show the data for the pristine and modified filters. The whole measurement time of 60 min is presented on the left side. The natural decay curve without the air cleaner is marked in blue, whereas the total decay curve with the air cleaner switched on at t = 0 is marked in red.

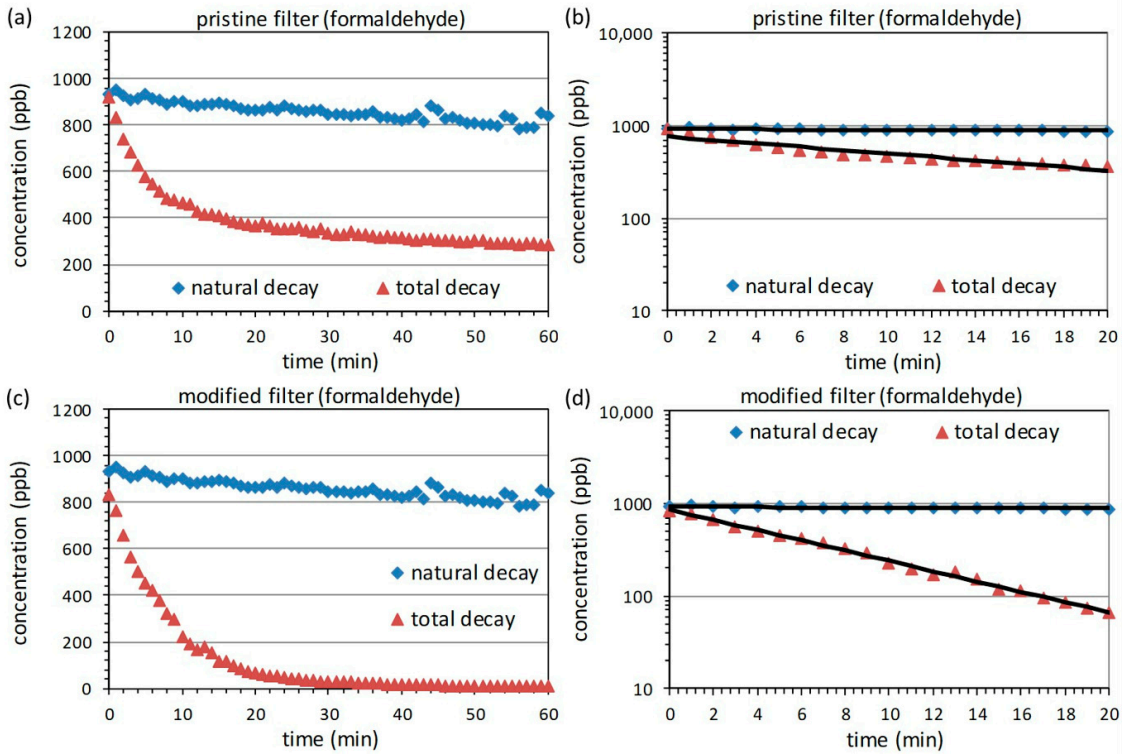

Figure 2. Natural (blue) and total (red) decay curves measured with formaldehyde for the pristine (**a**,**b**) and modified (**c**,**d**) filters. The whole measurement over 60 min is shown in the linear representation on the left side (**a**,**c**), whereas the first 20 min used for fitting are shown in the logarithmic representation on the right side (**b**,**d**). Black lines are exponential fits to the data points.

On the right side of Figure 2, the first 20 min of the data are shown in a logarithmic representation. Only those were considered for fitting since the decay curves started to deviate from the simple exponential approach in Equation (1) for longer measurement times. This was especially obvious for the pristine filter, for which the formaldehyde concentration did not approach zero but a stable equilibrium state between about 200 ppb and 300 ppb. This confirmed that the removal of formaldehyde by the pristine AC was due to a relatively weak physisorption. However, for the first 20 min, the decay curves followed the exponential model to a good approximation. as can be seen by comparing the

black lines with the data points. The values for the correlation coefficient R^2 fulfilled the minimum requirement of 0.90 of GB/T 18801:2015 [49] in all cases.

The derived values for the natural decay rate, the total decay rate, the correlation coefficient of the total decay rate, and the CADR calculated according to Equation (2) are listed in Table 1. Additionally, the adsorption efficiency was estimated on the basis of the estimated flow rates determined in the previous chapter. The natural decay rate was identical in both cases, as the same reference measurement was used. The data show that the CADR for formaldehyde increased from 71 m^3h^{-1} for the pristine filter to 219 m^3h^{-1} for the modified filter, which is close to the value of 220 m^3h^{-1} given by the manufacturer. This is a relative change in the CADR of more than 200%, which verifies that the modification had a substantial positive effect on the adsorption of formaldehyde.

Table 1. Natural and total decay rates, correlation coefficient for the total decay, derived CADR, and estimated adsorption efficiency for the two investigated filter types measured with formaldehyde.

Filter	$k_{nat}(min^{-1})$	$k_{tot}(min^{-1})$	R^2_{tot}	CADR(m^3h^{-1})	Adsorption Efficiency
Pristine	0.0037	0.0438	0.9081	71	~21%
Modified	0.0037	0.1280	0.9915	219	~62%

3.3. Measurements with Toluene

Figure 3 shows the decay curves measured with toluene for the pristine and modified filter in the same style as Figure 2 for formaldehyde. Again, a good agreement between the exponential fits and the measured data points was found for the first 20 min. The derived fit parameters and CADR values are listed in Table 2. In contrast to the previously found improvement for formaldehyde, a substantial decrease in the CADR from 289 m^3h^{-1} to 165 m^3h^{-1} was observed in the case of toluene. This corresponds to a relative reduction of 42%. Obviously, the positive effect for formaldehyde was accompanied by a negative effect for toluene.

Table 2. Natural and total decay rates, correlation coefficient for the total decay, derived CADR, and estimated adsorption efficiency for the two investigated filter types measured with toluene.

Filter	$k_{nat}(min^{-1})$	$k_{tot}(min^{-1})$	R^2_{tot}	CADR(m^3h^{-1})	Adsorption Efficiency
Pristine	0.0016	0.1661	0.9924	289	~85%
Modified	0.0016	0.0956	0.9952	165	~46%

3.4. Measurements with Nitrogen Dioxide

To complete the picture, Figure 4 shows the decay curves measured with NO_2 for both filter types in the same style as the previous figures. We note that during the measurements, a slight increase in the NO concentration in the chamber of about 20 ppb within 60 min was observed. That indicates that NO_2 was not only adsorbed by the AC but also partially catalytically reduced to NO, which is a well-known effect [59–61]. Therefore, the derived CADR is a slight overestimation of the real cleaning effect, as the NO can later oxidize back to NO_2. The NO_2 decay curves of both filters followed the exponential model over the first 20 min so that the parameters listed in Table 3 could be derived. As with toluene, a reduction in the CADR from 308 m^3h^{-1} to 239 m^3h^{-1} by the optimization was found with NO_2. However, the relative effect of a 22% reduction was less pronounced than that in the case of toluene.

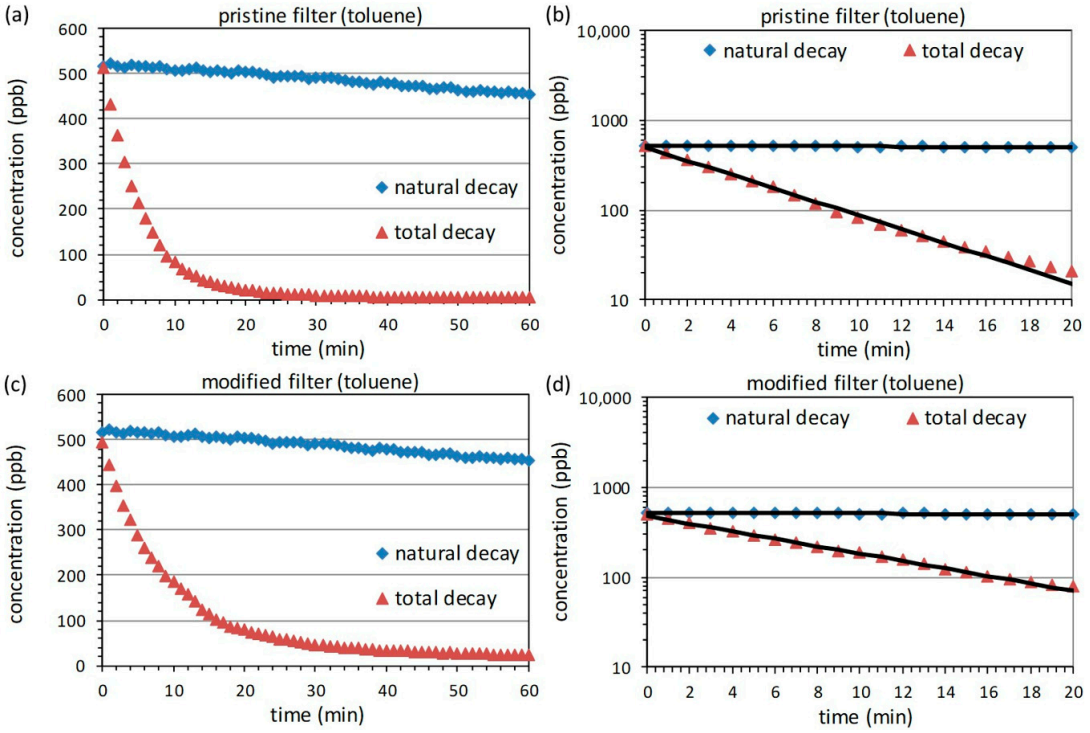

Figure 3. Natural (blue) and total (red) decay curves measured with toluene for the pristine (**a**,**b**) and modified (**c**,**d**) filters. The whole measurement over 60 min is shown in a linear representation on the left side (**a**,**c**), whereas the first 20 min used for fitting are shown in a logarithmic representation on the right side (**b**,**d**). Black lines are exponential fits to the data points.

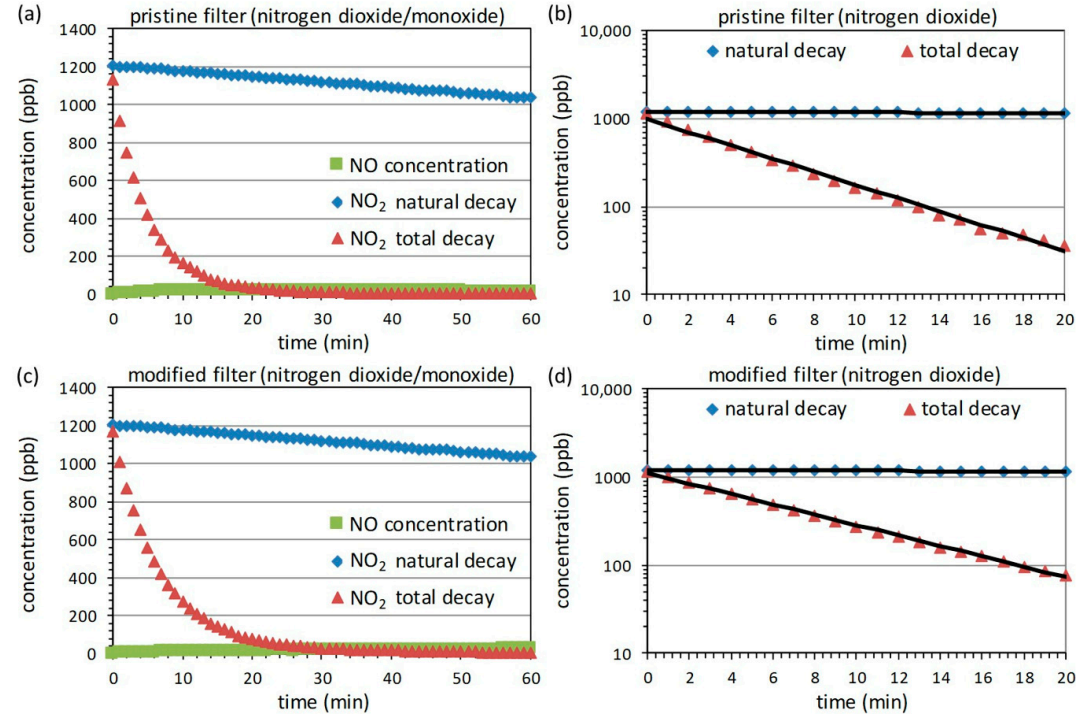

Figure 4. Natural (blue) and total (red) decay curves measured with NO_2 for the pristine (**a**,**b**) and modified (**c**,**d**) filters. Additionally, the measured NO concentration is shown in green for both filters. The whole measurement over 60 min is shown in the linear representation on the left side (**a**,**c**), whereas the first 20 min used for fitting are shown in the logarithmic representation on the right side (**b**,**d**). Black lines are exponential fits to the data points.

Table 3. Natural and total decay rates, correlation coefficient for the total decay, derived CADR, and estimated adsorption efficiency for the two investigated filter types measured with NO_2.

Filter	$k_{nat}(min^{-1})$	$k_{tot}(min^{-1})$	R^2_{tot}	CADR(m^3h^{-1})	Adsorption Efficiency
Pristine	0.0024	0.1773	0.9958	308	~90%
Modified	0.0024	0.1382	0.9987	239	~67%

4. Discussion

The results of the measurements with three different gases on two different filters are summarized in Table 4. The data show that the strong improvement in the CADR for formaldehyde was accompanied by a reduction for toluene and nitrogen dioxide. As the type of modification of the AC is not known for confidentiality reasons, the exact cause for this competing effect could not be identified. Nevertheless, we discuss several potential explanations below that might be transferrable to other cases in which filters are optimized for the removal of formaldehyde.

Table 4. CADR of the two investigated filter types for three different gases.

Parameter	Formaldehyde	Toluene	Nitrogen Dioxide
CADR pristine filter	71 m^3h^{-1}	289 m^3h^{-1}	308 m^3h^{-1}
CADR modified filter	219 m^3h^{-1}	165 m^3h^{-1}	239 m^3h^{-1}
Change with modification	+208%	−42%	−22%

The first potential explanation is that a change in the chemical surface properties induced by the modification could lead to a reduced efficiency for toluene and NO_2. For example, increasing the polarity of AC is known to decrease the performance for non-polar VOCs, such as benzene, toluene, ethylbenzene, and xylene (BTEX) [62]. However, the impregnation with amines typically used to improve formaldehyde adsorption is also known to lead to better adsorption of NO_2 [63,64]. As this is in contradiction to the findings here, a change in the chemical surface properties does not seem to be the dominant effect.

Second, it is known that the impregnation of AC can also have a negative influence on the physical properties, such as the Brunauer–Emmett–Teller (BET) surface area as well as the pore volume and size distribution [65]. If this had been the case here, it would in principle lead to a reduced adsorption efficiency for all three adsorbates [66]. However, if the reduction can be overcompensated by the improved chemical surface properties for the adsorption of formaldehyde, it is plausible that a reduction in the CADR is only observed for toluene and NO_2.

A third potential explanation is that the AC is partially replaced by another adsorbant. For example, blends or composites of AC with zeolites [67,68], mesoporous silica [69], silica aerogels [70], or metal oxides [71] have been used in the past to optimize the adsorption efficiency for specific gas mixtures. However, if the added adsorbant is more efficient for formaldehyde but less efficient for toluene and NO_2 than the original AC, the degradation for the latter gases could be simply explained by a lower amount of AC integrated into the filter.

5. Conclusions

The results show that, for an exemplary air cleaner, the optimization for formaldehyde by the modification of the AC filter can have detrimental effects on the adsorption of other gases, such as toluene or NO_2. This leads to the conclusion that testing standards for air cleaners should not only consider formaldehyde but also other gases to reveal such potential negative side effects. This concept was recently followed in the latest revision of GB/T 18801:2015 [49] to GB/T 18801:2022 [72], which now also provides a measurement with a gas mixture of formaldehyde, toluene, butyl acetate, and styrene as an option. Similarly, the recently published US testing standard AHAM AC-4:2022 [73] suggested tests with formaldehyde, ammonia, toluene, n-heptane, and d-limonene. In addition, the new

international testing standard IEC 63086-2-2 [74], which is currently under development, plans to make more than a single gas mandatory for testing.

Should such an extended test program reveal drawbacks of optimization, a possible solution could be to use blends of different adsorbants that complement each other with the aim of finding the best compromise for a typical composition of indoor air pollutants. Alternatively, novel approaches for surface modifications to AC, e.g., by the integration of specific nanoparticles [75–77], might show less negative impacts on the adsorption of other gases. However, this needs to be demonstrated in future studies.

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