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Reflections on the efficiency and toxicological implications of NTP air cleaning devices

Abstract

By means of electrical discharge between two electrodes a special type of air cleaning devices generates a non-thermal plasma (NTP) consisting of particles of high energy, which are supposed to react with air pollutants to form toxicologically harmless products. Theoretical considerations and literature data justify doubts about attainable complete oxidation of organic indoor air pollutants by this technology. The generation of ozone caused in this process is inevitable and its release probable. According to the information available a sound hazard assessment according to the German Act on Occupational Health and Safety (Arbeitsschutzgesetz) and the Ordinance on hazardous substances of 2004 is not possible. Therefore, the operation of such devices in direct proximity of persons cannot be recommended.

1 Introduction

The type and concentration of air pollutants in industrial waste air flows are usually known, which means that the cleaning system can be tailored to the specific conditions. By comparison, the situation for universal cleaning systems for indoor air is entirely different. Their task is to reliably remove a multitude of different substances which are present in much lower concentrations than in industrial emissions (order of magnitude ppb to 1 ppm).

The remedy to this problem is promised by manufacturers of a special type of air cleaning device that makes use of reactive oxygen species (ROS) and is said to ensure the elimination of pollutants by oxidative conversion into safe reaction products, such as carbon dioxide and water. Further, their makers also claim that such devices kill microorganisms and additionally enhance the sense of well-being of persons in the room. In the trade, this technology is marketed under a variety of terms, such as plasma field ionisers/generators, NTP generators/ionisers and also air ionisers.

In the following we wish to theoretically assess whether the stated goals are capable of achievement technologically and toxicologically and from the point of view of occupational medicine.

1.1 Mode of operation of NTP reactors

The various air cleaning devices available on the market (referred to in the following as NTP reactors) make use of a technology that has its origin in the Siemens ozoniser. They generate so-called non-thermal plasma (NTP) by releasing electrons of high kinetic energy – about 3 to 6 eV corresponding to 290 to 580 kJ/mol – by electrical discharge between two electrodes and accelerating them in the electrical field (**Figure 1**). Depending on the design, direct or alternating voltage is applied continuously or in pulses to concentric, plate- or rod-shaped electrodes. By means of various methods, e.g. by corona or dielectric barrier discharge, spark discharge is prevented. The physical principles of the generation of NTP in barrier discharge reactors have recently been described in detail by *Chirokov* et al. [1].

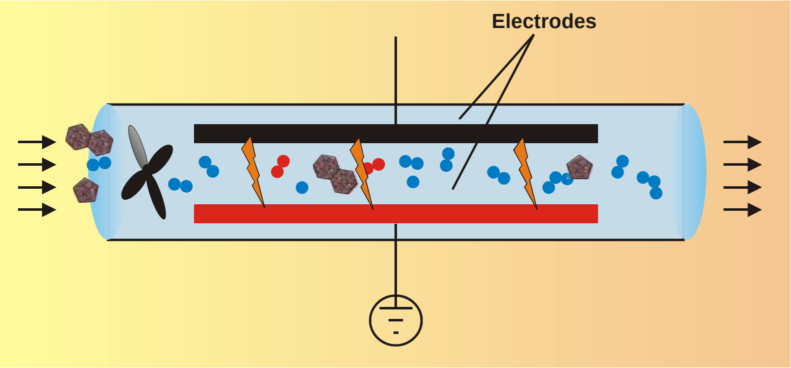

Figure 1:
Diagram of an NTP reactor

A ventilation system guides the pollutant-laden air through the reaction zone between the electrodes. The energy of the electrons is now exploited to activate the atmospheric oxygen contained in this air flow by forming ROS that should be capable of eliminating the pollutants present in the carrier gas by oxidation. Some commercially available devices are equipped with filters, catalysts and/or additional ion sources.

At first sight this method of air cleaning would appear simple in principle in terms of technology and effect, yet the various scenarios resulting from the application of NTP are complex. In fact, account has to be taken of not only “pure” air – 78.08% nitrogen, 20.95% oxygen, 0.93% argon, 0.03% carbon dioxide, trace gases and water in changing proportions – and specific air impurities, but also the typical organic air constituents. These are usually also present in “unpolluted” interiors and include d-limonene, camphene, ethanol, formaldehyde, acetaldehyde, isoprene, styrene, glycol ether and various aromatics.

1.2 NTP reactors for indoor use

We wish to deal with the use of NTP reactors as indoor air cleaners from three points of view:

- **Generation of primary products in non-thermal plasma**

Manufacturers’ product descriptions contain conflicting information on the reactive oxygen species (ROS) arising in NTP. Thus it is initially important to review the compounds that may form and how they arise. We therefore have to estimate whether and which ROS can arise during operation in pure air. Another question to be considered is the quantities in which the generated ROS are introduced into the indoor air. At the same time, air impurities may also be directly attacked by the NTP and released again in the form of reaction products.

- **Reactions of the formed oxygen species with air impurities**

Since NTP reactors are available for universal use for the removal of unwanted air constituents, a very general review of the reaction paths of such air impurities with the generated ROS is necessary. This should always be conducted in the light of the desired goal of harmlessly eliminating air impurities with the aid of NTP reactors.

- **Health issues**

The NTP reactors reviewed here only perform their task without reservation when they convert pollutants in the indoor air with the generated reactive species as quantitatively as possible into harmless substances. The observance of toxicological effect thresholds is universally demanded, and not only in terms of the air pollutants and their degradation products, but also with respect to the reactive substances arising directly during the operation of air cleaning systems.

2 Primary products in non-thermal plasma

2.1 Atomic oxygen and ozone

As already indicated, NTP generators work on the principle of the Siemens ozoniser. To generate ozone (O_3), they initially break down a molecule of dioxygen (O_2) into atomic oxygen. The high energy required for this (5.16 eV corresponding to roughly 500 kJ/mol [2]) comes from electrical discharge (see section 1.1). The individual oxygen atoms, an extremely reactive species, can now react with a further molecule of dioxygen to form ozone (**Figure 2**). The second reaction in Figure 2 explains the reactivity of ozone, as it can very readily serve as a source of atomic oxygen by reversing this process.

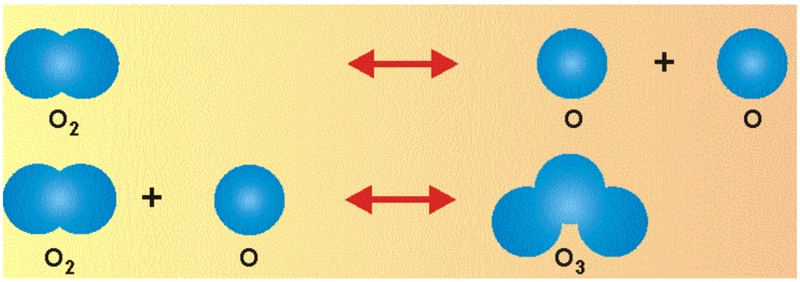

Figure 2: Generation of ROS from oxygen

Although the energy input in NTP reactors is much lower than that of ozonisers, the emergence of ozone is inevitable as a result of its working principle[1]. No general statement can be made about the extent in each case, since this depends on the particular device’s design and performance. In view of the relatively long life of ozone (**Table**), its release in ranges outside the plasma zone is plausible.

On the other hand, atomic oxygen, because of its reactivity, has an extremely short half-life (Table) and release is not therefore expected.

Table: Half-lives of a number of reactive species

Species	**Life**	**Literature**
O	0.1 s	[3]
·OH	1 s	[4]
·OOH	1 min	[4]
O_2 ($^1\Delta_g$)	< 64 min	[5]
O_2^-	s to min	[6; 7]
O_3	1 month	[4]
H	Extremely short	[4]
N	Extremely short	[4]
NO	1 day	[4]
NO_2	Several days	[4]
HNO_3	Several days	[4]

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[1] The energy input conforms to a Boltzmann distribution.

2.2 Singlet oxygen

In connection with NTP reactors, there is often mention of singlet oxygen as the reagent[2]. This is the term given to a form of dioxygen that changes into a more reactive state by absorbing energy (**Figure 3**). Although the quantity of energy required for this (0.98 eV corresponding to 94.3 kJ/mol [2]) is not particularly large, this form of excitation is tied to special conditions. The selection rules, which are explained by theoretical chemistry, make this activation unlikely without a “sensitiser”. In strong electrical fields as present in NTP reactors, such special conditions are in fact present. There is no strong evidence in the existing literature that this can weaken the selection rules. It therefore remains unclear whether singlet oxygen really is generated to an appreciable degree.

2.3 Hydroxyl radical (·OH)

The hydroxyl radical arises due to the reaction of atomic oxygen with ubiquitous water (H_2O, **Figure 4**) and also as a possible product of ozone and unsaturated compounds (section 3.2). It is characterised by its relative reactivity with comparably high stability and the thus associated life of about a second (Table 1). Its release, albeit minimal, from the reaction chamber of an NTP reactor cannot therefore be excluded.

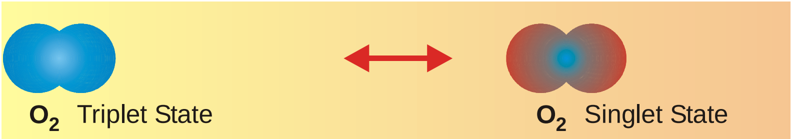

Figure 3: Excitation of dioxygen

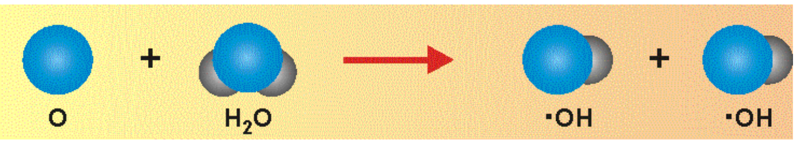

Figure 4: Occurrence of the hydroxyl radical

2.4 Further intermediates

Electrons released during electrical discharge can encourage the reduction of the dioxygen to yield the superoxide radical anion (O_2^-). The presence of water yields the formation of the hydroperoxyl radical (·OOH, **Figure 5**). The breakdown of water molecules to form hydrogen atoms and hydroxyl radicals is a further source of reactive species. By reacting with oxygen or ozone, both can generate the hydroperoxyl radical (·OOH, **Figure 6**).

Air consists of 78% nitrogen which, like dioxygen, has the diatomic form as an N_2 molecule. The dissociation of N_2 yields atomic nitrogen. This, however, requires much higher energy than the equivalent reaction of the O_2 molecule and can only therefore be expected to a very limited degree, if at all, in NTP reactors.

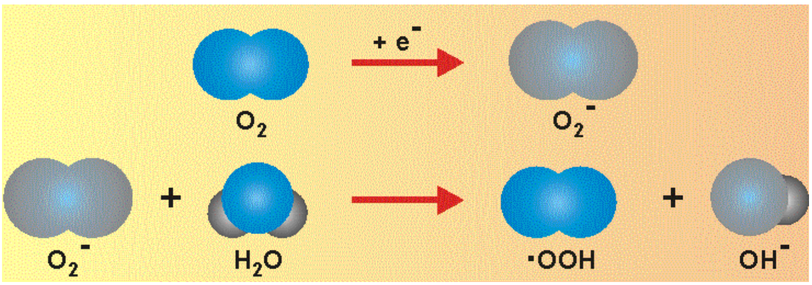

Figure 5: Occurrence of the superoxide radical anion and its reaction with water

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[2] The chemically correct designation is O_2 ($^1\Delta_g$).

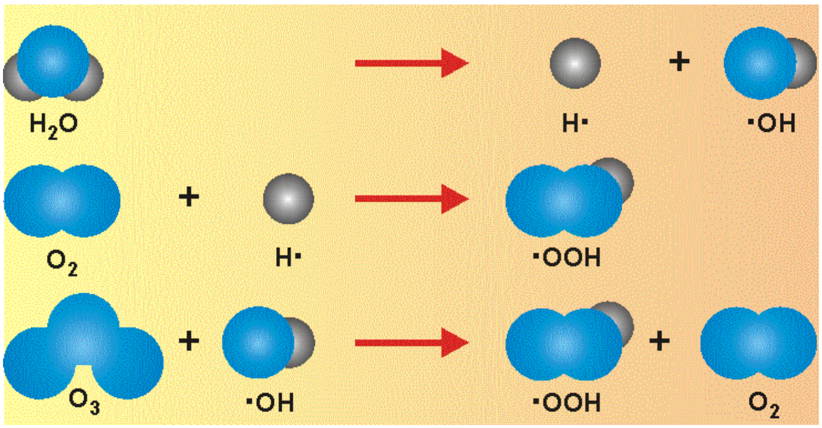

Figure 6:
Further ways in which the hydroperoxyl radical can occur

2.5 Direct reactions of organic air constituents with non-thermal plasma

Electrical discharges in discharge reactors are basically capable of initiating direct reactions of the organic air impurities. Depending on their type, these can be converted by electron uptake into radical anions. This activates them for subsequent reactions with the other substances present, such as ROS or other air constituents, or they stabilise themselves by fragmentation. Much the same applies here to direct conversions with ROS: A multitude of different products is conceivable, and in the event of fragmentation even an increase in the pollutant concentration is possible. The extent depends again on the performance and characteristics of the NTP reactor and cannot be estimated without farther-reaching investigations.

2.6 Digression on “air ions”

A special group of NTP reactors (air ionisers) promises an enhancement of well-being and improvement in health by generating negative ions. Furthermore, it is also argued that aerosol particles contained in the indoor air form condensation nuclei by electrical charging and these nuclei precipitate faster after their agglomeration. This leads to a reduction in the particulate concentration, although an appreciable effect depends very strongly on the performance of the device used and room size. There are nevertheless studies that cast doubt on a marked or lasting effect on both counts [6; 8].

Air ionisers operate with a relatively low energy input. All the same, studies have already shown that ozone is released even when these devices are used [9]. From the point of view of occupational health, there would not appear to be a need to assess these devices and their purpose separately. Depending on energy input, the same applies here as to NTP reactors.

3 Reactions of reactive oxygen species with organic air impurities

3.1 Atomic oxygen

Atomic oxygen is one of the most aggressive oxidants. With its help, it would be possible to fully convert organic air pollutants (Volatile Organic Compounds = VOCs) into simple compounds such as carbon dioxide and water, provided enough of it is available. Heteroatoms such as sulphur and nitrogen, which may be contained in VOCs, would then inevitably result in the formation of sulphur and nitrogen oxides.

At the same time, the generation of large quantities of atomic oxygen in NTP reactors is not desirable because of the similarly large quantities of ozone that would arise (Figure 2) [10]. Because of the enormous excess of dioxygen in relation to possible air impurities (21% O_2 compared to < 1 ppm air impurities), small concentrations would mainly combine with atomic oxygen to form ozone. This means that air pollutants can be decomposed by direct reaction with atomic oxygen to yield at best partially oxidised products. In unspecific conversions, these can result in new compounds.

3.2 Ozone

Reactions of ozone with organic compounds have been frequently described in the literature. In synthesis, ozone is used partly because it is useful for selective conversion in the form of so-called ozonolysis. This means that with the aid of ozone certain specific functional groups of organic molecules can be modified without attacking the rest of the molecule. If the reactions with ozone are uncontrolled (as in an NTP reactor), these functional groups are also preferentially attacked. Unsaturated hydrocarbons, for example, yield via ozonides and Criegee intermediates – in some cases by breaking down the initial molecule – unspecifically different compound groups such as carboxylic acids, aldehydes, ketones and hydroperoxides. Hydroperoxides of the type $R_2C{=}CR{-}OOH$ (from a *Criegee* intermediate) decompose further, releasing the ROS hydroxyl radical [2]. With ozone concentrations in the low ppm to high ppb range, the quantitative conversion of organic air pollutants into CO_2 and H_2O cannot be expected [11].

3.3 Singlet oxygen

If singlet oxygen is formed in appreciable quantities, it would also appear to be of only limited use as an air purifier. It is worth noting that it is used in preparative chemistry not as a universal but as a selective oxidant for the controlled modification of unsaturated compounds. In NTP the associated reaction products can be expected, e.g. peroxo compounds, possibly followed by rearrangements. This conflicts with the envisaged complete breakdown of a wide range of air pollutants.

3.4 Hydroxyl radical

Of the remaining ROS, the hydroxyl radical (·OH) is the most important. It is highly reactive and attacks organic air pollutants such as unsaturated compounds, aromatics and aldehydes [12]. A wide range of products can therefore be expected (alkyl radicals R·, alkoxyl radicals RO·, alkyl peroxyl radicals ROO·). Farther-reaching or complete breakdown of the primary products is improbable because of the small quantities involved and the almost complete lack of subsequent photochemical reactions in interiors.

3.5 Nitrogen oxide compounds

As already suggested above, NTP reactors are unlikely to encourage the formation of nitrogen oxides from atmospheric oxygen in any appreciable quantities. Nevertheless, up to an average of 100 ppb of nitrogen monoxide (NO) is possible in interiors as a consequence of harmonisation with outdoor air concentrations [13]. To this extent, nitrogen dioxide (NO_2) from NO and atomic oxygen, ozone or hydroperoxide (·OOH) can thus be assumed to form (**Figure 7**).

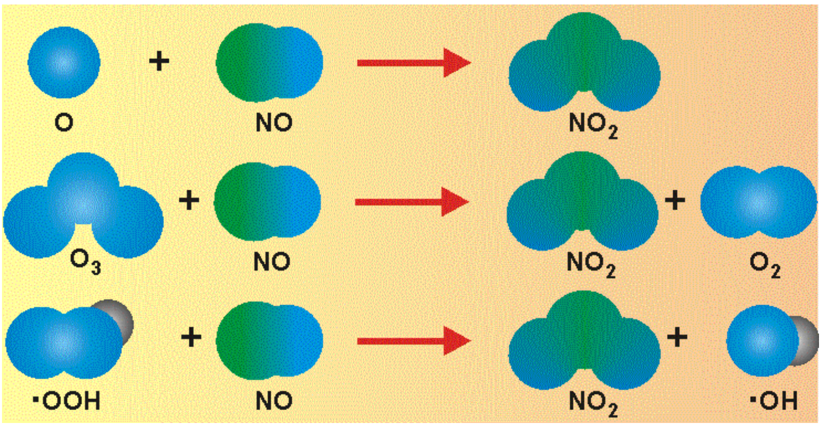

Figure 7: Reactions of nitrogen monoxide

In the last reaction in Figure 7, an ROS is generated in turn with the hydroxyl radical. Further subsequent products are then nitrogen trioxide (NO_3) and dinitrogen pentoxide (N_2O_5), from which nitric acid can form with water (from humidity in the air) or with VOCs (**Figure 8**).

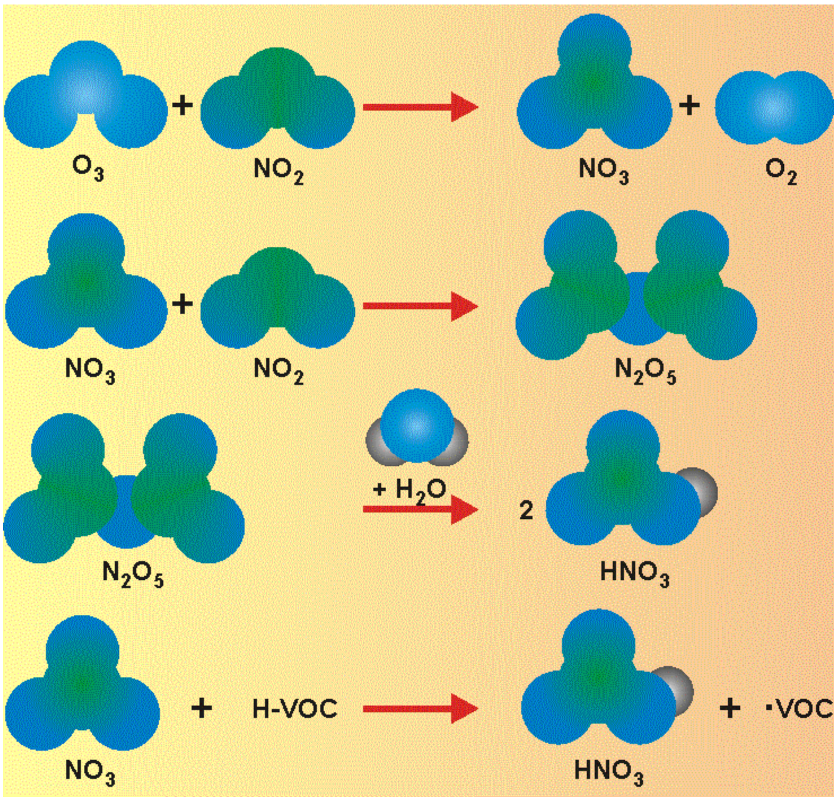

Figure 8: Further reactions of nitrogen compounds

4 Health issues

4.1 Ozone

There is no let-up in the discussion of the health effects of ozone and “safe” exposure concentrations. We therefore refer to the relevant literature on the toxicity of this substance [10; 14 to 17].

To protect human health from ground-level ozone, the 33rd Ordinance for the Implementation of the German Federal Immission Control Act [18] specifies a target value of 120 µg/m³ as the maximum 8-hour average in the course of a day with 25 transgressions permitted per calendar year. The information and alert thresholds for ground-level ozone are 180 and 240 µg/m³ respectively as 1-hour averages. The Swiss Clean Air Act (LRV) is even stricter in specifying that 120 µg/m³ as the 1-hour average may only be exceeded at most once per year [19].

The workplace air limit value for ozone applicable in Germany according to TRGS 900 until the end of 2004 was set at 0.2 mg/m³ with reference to an older maximum workplace concentration (MAK) value of the Commission for the Investigation of Health Hazards of Chemical Compounds in the Work Area, the so-called “MAK Commission” of the German Research Foundation (DFG). However, back in 1995, it was the very same MAK Commission that had suspended this MAK value because of uncertainty about its effect threshold and suspected carcinogenicity [14]. For this reason alone, the installation of an ozone source in frequented interiors is ill-advised. In concentration measurements conducted by the BG for the Construction Industry in model rooms in which NTP reactors of different makes were operated, the then valid workplace air limit value of 0.2 mg/m³ was exceeded near to the expulsion vent [20]. The precise test conditions and measurement results are to be published here in the near future.

4.2 Nitrogen compounds

Much like the equivalent value for ozone, the MAK value for nitrogen dioxide was suspended by the DFG Commission because of its suspected carcinogenicity. The Swiss pollution control value for NO_2, a 24-hour average that may only be exceeded once per year, is 80 µg/m³ [19]. The Indoor Air Hygiene Commission (IRK) of the German Federal Environment Agency (UBA) has set guide values for NO_2 in indoor air of 350 µg/m³ (0.5 hours) as the short-term exposure limit (RW II K) and 60 µg/m³ (weekly average) as the long-term exposure limit (RW II L). Guide value II is the concentration of a substance that calls for immediate action as soon as it is reached or exceeded, as this concentration is likely to pose a health risk particularly for sensitive persons present in these rooms for extended periods [21]. However, since these values are often exceeded solely as a result of entrainment from the outdoor air, the exclusion of further emission sources is important. Considerable concentrations of nitrogen monoxide are not expected in view of the oxidative conditions in rooms in which NTP reactors are operated as long as the guide values for NO_2 are complied with. There are no suitable studies for assessing the risk of inhaling nitric acid (HNO_3) in the low ppb range.

4.3 Degradation products of air constituents

As exhaustively explained, the products of reactions of organic air constituents with ozone, singlet oxygen or the hydroxyl radical are by no means exclusively CO_2 and H_2O. In fact, a cocktail of new substances is generated, including aldehydes and organic radical compounds whose irritant effect has been extensively documented. If the initial molecule is broken down, an increase in the particulate concentration may even result.

Tests with relatively high concentrations of model pollutants (at least 15 ppm) in NTP reactors yielded a large range of incompletely degraded reaction products. Depending on the initial substance, the substances detected include carbon monoxide, hydrocarbons, aldehydes and organic and inorganic acids [22 to 25]. A reactor with a concentric electrode arrangement converted 160 ppm of trichloroethylene into a product mixture containing dichloroacetyl chloride, phosgene and hydrogen chloride [26]. It would be going too far at this point to explain in detail the toxicology of individual products, particularly since the emitted quantities, reflecting the low initial concentrations of indoor air contaminants, ought normally to be low.

Since it is impossible to estimate the synergistic health-hazardous effects of these various compounds, they cannot be entirely excluded. In this connection, we wish to draw attention to a study with probands exposed in an exposure chamber to concentrations of about 0.05 mg/m³ of ozone and about 0.1 mg/m³ of formaldehyde. The irritant effects described in this study were ascribed to the combined action of the two substances which had not been observed during exposure to one of the two substances by itself. It is interesting to note that a large portion of the formaldehyde was generated by ozonolysis of the room's own pollutant emissions [27].

5 Conclusion

The idea of using oxygen for the removal of organic indoor air impurities for the benefit of health and the environment looks at first sight useful and promising. However, of the species formed in non-thermal plasma, only atomic oxygen would probably be capable of performing this ambitious task. Producing this in large quantities, would cause the formation of huge quantities of ozone as a result of subsequent reactions with molecular oxygen, as the ratio of its competing reagents O_2 and VOCs is greater than 210,000 : 1 (i.e. 21% O_2 compared to < 1 ppm VOCs). This means that ozone would be inevitable as the main product. What's more, the heteroatoms contained in VOCs, such as nitrogen and sulphur, would be released as oxides.

In industry, the systems operated for the decontamination of exhaust air make use of NTP technology merely to bring pollutant concentrations in a high ppm range to below the emission limits. They are, however, usually equipped with filters and catalysts.

After evaluation of all the available facts, it can be concluded that it is not possible to adapt NTP reactors to the cleaning of weakly contaminated indoor air without further ado. A high input of energy into the system yields unacceptable ROS concentrations, while a low input yields partially oxidised products whose irritant effect is in some cases stronger than that of their precursors. At the same time, the release of ROS, and particularly ozone, can also be expected in the latter case. This forecast based on theoretical considerations has been confirmed by independent guide measurements.

On the basis of our experience, the manufacturer information on NTP reactors for indoor use is insufficient in terms of the actual air cleaning capacity (including deodorisation and the killing of microorganisms) and possible effects on human health. In isolated cases, compliance is claimed with the ozone MAK value, which is no longer applicable in Germany; no valid documentation from neutral bodies is available. Despite great efforts on our part, informative publications that identify and quantify possible products of reaction under real indoor air conditions have not come to our knowledge. This means that a sound risk assessment under the terms of the German Occupational Safety Act (Arbeitsschutzgesetz) and the new Hazardous Substances Ordinance (Gefahrstoffverordnung) is not possible. The use of NTP reactors therefore cannot be recommended. Furthermore, we must warn against the operation of such reactors in rooms frequented by people.

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