

Fragments of Disaster Medicine IV

Radicals in our nature and the nature of radicals

Summary:

In our industrialised society, people of all ages are increasingly suffering from recurrent allergies and inflammations. Thus, despite our high-performance medicine, intrinsic (i.e. non-allergic and often infection-related) asthma and obstructive pulmonary disease (COPD) are currently strongly represented with their considerably higher mortality. What is the contribution of air and environmental pollution? How does global warming affect our health with foreseeable catastrophic scenarios? Risk factors and triggers of inflammatory processes are bacteria, well-known but also novel, pandemic viruses.

In order to understand inflammation, newer biological-chemical principles must be included: Reactive free radicals will play a role. They are called "oxidants" when they dock to a molecule as an "antioxidant", take up an electron or split off one of its atoms. Small reactive radicals move in a diffusion-controlled manner and also attack biological molecules at their destination. Rapid, damaging exchange reactions can often be repaired in tissue. However, if the aggressive oxidative particles outweigh the mechanisms of antioxidative compensation, inflammation develops.

Reduction and oxidation are not equally likely locally. In some molecules, there are segments that are preferred for radical reactions as "functional groups". At these, free radicals can dimerise to less reactive molecules or form temporarily larger adducts. In order to intercept reactive species at the beginning of severe inflammatory processes, functional groups can be used medicinally. - Why is this often not recognised? - Natural science subjects have so far been dominated by older ideas about chemical bonding. In these, polar to ionic structures dominate *with continuous* transitions. The filling and emptying of the outer electron shells of reactants according to a noble gas configuration (octet rule) is part of the teaching. Free radicals have no place in this thinking. But the bonding of atoms in molecules solely from heteropolar, electrostatic salt structures and the theory of their electrolytic dissociation in solution is not compatible with quantum theoretical calculations.

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Abstract:

Some knowledge of chemistry seems necessary again to understand the fragments of disaster medicine. - A special case for the connection of the two atoms A and B is the heteropolar ionic compound A^+B^- . Its electrolytic dissociation in solution was established by S. Arrhenius and its crystalline structure was perfected by W. Kossel with the octet rule, that means: In polar bonds atoms try to arrange their outer bond electrons similar to outer noble gas shells. On the other hand, however, a covalent bond consisting of atoms A and B is verified by quantum chemical calculations, even if this bond in A-B showed a certain polarity before. Each bond break yields uncharged fragments of reactive free radicals. They are fundamental to understanding of becoming and decay, diseases, pandemics, catastrophes and the recognition of changes in the environment, in the atmosphere as well as global warming.

Kossel's binding theory had a strong influence on the development of chemistry in Germany. However, it does not provide a space for the existence of free radicals, nor does it provide an adequate environment for homolysis and homo-synthesis in the world of biology. Moreover, Kossel's theory collocated polar and ionic structure from experimental synthetic chemistry into physiological processes, which are exactly impossible there for thermodynamic reasons.

Headwords:

functional groups - free radical reactions - medical significance - inflammation - oxidants and antioxidants - environmental pollution - global warming explanation - Svante Arrhenius: electrolytic dissociation - Wilhelm Ostwald: nitric acid synthesis starting from ammoniac. Walther Kossel: chemical binding theory based on ionic bond and octet rule - **Arrhenius-Kossel's dilemma** - water treatment and disinfection - new dangerous germs - from endemic to pandemic diseases - prophylaxis and/or therapy.

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Foreword: Healthy living in a sick environment? - Understanding and preventing inflammation - Radicals in view: an aid to medicine

If homolytic forces act on biological-chemical molecular bonds in the tissue, smaller parts can be created. These are radicals. What is the nature of these forces? Does radiation, heat, pressure have an effect? Do gaseous or liquid radicals react via impact? What do these particles mean for our health today and in the future?

Radicals are fractions of chemical structures with at least one unbound electron ¹. They dimerise or spontaneously bind comparable particles. Thus, O₃ molecules of the "ozone layer" (corresponding to the lower stratosphere at an altitude of approx. 15 to 25 km) are converted to chlorine oxide radicals (ClO•) and molecular oxygen (O₂) by chlorine atoms (Cl•), which are split off from chlorofluorocarbons (CFCs) due to UVB radiation. The ClO• itself degrades ozone to O₂ and again catalytically active chlorine (→Cl•). The "lost" O₃ molecules then offer no further protection from the extremely dangerous UV radiation of the sun ². An enlargement of the "ozone hole" would have globally catastrophic consequences ³.

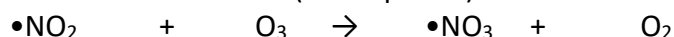
Radicals are involved in the development of many diseases ⁴. Although indispensable in the life-sustaining flow equilibria of physiological metabolism ⁵, these are the focus of genetic defects ⁶⁻⁸. Formation and elimination of radicals always require critical analysis in a variety of different reactions on the part of chemistry, biology and medicine.

•NO and •NO₂ are formed in the troposphere from nitrogen oxides •NO_x from traffic, industry and households. Involved in ground-level ozone formation, they were therefore considered environmental noxious substances for humans and indirectly for plants ^{9,10}. This was experimentally disproved for •NO in 2016 ¹¹. Like ozone, many radicals are inflammatory mediators.

1. Separation of chemical bonds into radicals

The extremely aggressive hydroxyl radicals (HO•) propagate inflammation, often triggered by viruses and/or bacteria. In biology, neutral to slightly acidic pH values correspond to a microenvironment characterised by inflammation. Radicals are allegedly involved in the development of cancer and ageing ^{12,13}. On the other hand, reactive species in the "respiratory burst" ¹⁴ are essential tools of migratory cells (e.g. macrophages) to destroy viruses, bacteria, fungi and protozoa. They are thus jointly responsible for maintaining the immunity of higher organisms.

Ecologically, HO• - radicals (in a recycling process with natural isoprene) are considered the most important purifier of atmospheric air pollutants ^{15,16}. Their molecular nature sometimes appears "dodgy": Human-toxic •NO₂ also acts as a greenhouse gas in very low concentrations, as it is able to reduce (atmospheric) ozone.



•NO₃ radicals, comparable to HO•- molecules, oxidatively degrade air pollutants and are a further purifying agent of the atmosphere ^{15,16}.

Research into sessile radicals on solid surfaces is in its infancy. - Chemists, biologists and physicians are primarily interested in oxidation- and reduction-effective species in tissues. Of concern are: *Reactivity* and *diffusivity*: *Reactive* radicals are very small. HO•, •NO or -•NO₂ lack the substituents hiding the "lone" electron (stereo-chemically). *Diffusivity*: Small, less polar radicals diffuse statistically to their reaction site in tissues.

The binding of radicals to biomolecules or antioxidants means reduction ¹⁷ for the latter. Reactions of the "scavengers" ¹⁷ must be examined closely, especially since radicals can also function as essential messengers ^{17,5} in metabolism. In the context of chemistry, biology, emergency and disaster medicine ¹⁸ molecular instabilities require an examination of both mobile and sessile radicals.

... 3.4.-Diiodo pyrrole ¹⁹ as a possible approach to homolysis

The 3.4-symmetrically iodinated molecule proves to be unstable both during synthesis and during storage (as a crystalline solid) in the presence of minute amounts of oxygen. Another characteristic is a considerable sensitivity to UV light. The sometimes violent, non-ionic decomposition (with the formation of black-violet vapours of iodine, carbon, N₂ or ammonia ^{*}) is favoured by intramolecular interactions of the voluminous substituents in positions 3 and 4 with H-atoms of the neighbouring positions 2 and 5 at increased ring tension of this heteroaromatic ²⁰. The lack of functional groups to protect against oxygen requires working under inert gas (Schlenk-Technik^{*}) if possible and storing the synthesised preparations at low temperatures protected from light. For the decomposition of 3.4-diiodo pyrrole, traces of bi-radical triplet oxygen are sufficient. The homolytic decomposition proceeds probably starting with the cleavage of the C3-C4 bond - apparently quite simple in formulation - but probably much more complex in reaction:

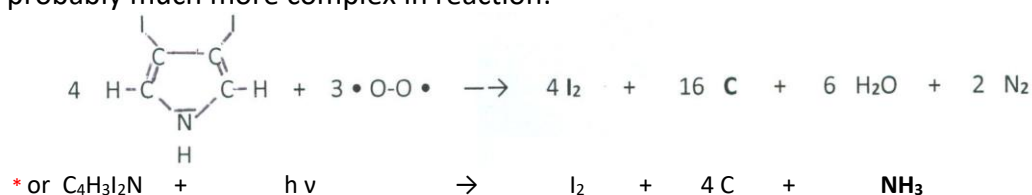


Fig. ① 3.4.-Diiodo pyrrole - possible radical induced decay modes

Are •OH radicals from H₂O₂ also initially involved in the decay as highly reactive species? Or do nascent carbon molecules and various reactive species from iodine and water in the gas phase complete the destruction of volatile 3.4-diiodo pyrrole? Hydroxyl radicals are probably involved in the decay. They could form from HOI. - But how stable is pyrrole as a building block of numerous biomolecules? - A ring system of tetra-pyrrole is a component of the porphyrin skeleton of blood dyes (haem) and chlorophyll (leaf green) ²¹. Four intact pyrrole particles can still be recognised in bile dyes (e.g. bilirubin, biliverdin) as degradation products of blood dyes ²¹. In the biomolecules concerned, the heteroaromatics centre free or bound metal atoms of the main group and transition elements via electrons of nitrogen with complex formation. In the process, the hydrogen atoms of the NH-functions of each of pyrrole forming stones are replaced by metal atoms. These in turn interact with the other pyrrole bodies. In both respiration and photosynthesis, radical reactions of complexes analogous to those of porphyrin are physiologically of vital importance. Pyrrole itself is obviously very stable in the complexes mentioned. An extensive nomenclature exists for tetra-pyrrole and its numerous derivatives ^{21,22}.

* Note: According to Wilhelm Schlenk (1879-1943) in German also "Schlenktechnik": Working under inert gas: i.e. exclusion of oxygen and water vapour, at chemical reactions. - Currently, argon is increasingly being used as an inert gas instead of nitrogen.

2. Functional groups: Reactions with free radicals

Example: Dimethyl sulfoxide (DMSO)

Until 1965, dimethyl sulfoxide (DMSO) was considered a highly effective drug against cancer and inflammation-related diseases ²³. However, the National Institute for Health and Care Excellence (NIHCE) found systemic and topical applications of this substance insufficiently documented, with one exception ^{24a, b}. However, in the "IFA GESTIS Substance Database" ²⁵ in accordance with an online publication (not discussing radicals!), pain-reducing and anti-inflammatory applications of DMSO are further registered with low toxicity ²⁶. - According to earlier biochemically motivated publications, the disposal of free radicals and reactive species is primarily based on effects of antioxidant enzymes such as catalases, glutathione peroxidases, superoxide dismutase, thioredoxin reductases, glutathione, coenzyme Q₁₀, antioxidant micronutrients such as vitamins A, C, E, carotenoids and food supplements ²⁷. Even *haematic* iron and iron-containing proteins in human metabolism have been postulated as "antioxidant" - but without description of chemical mechanisms ^{28,29}. Properties of direct radical scavengers ^{30,*} and corresponding "scavenger" mechanisms have not been discussed in medical publications to date. Presumably, DMSO (see below) belongs to a group of direct radical scavengers. According to the cited study from Belgium ²⁶ hydroxyl radicals (HO•) are supposed to be intercepted by means of local application of DMSO in arthritis via the "inhibition of inflammatory cytokines" (thus *again not directly!*). With regard to the course of radical removal, there are therefore no convincing and chemically recognised mechanisms either for this indication nor for previously described applications. But how is an understanding of the direct disposal of reactive species achieved?

Fictitious "boundary structures" are graphical aids in chemistry. They often allow the development of plausible models regarding a possible density distribution of the charge - as in DMSO. This particle corresponds to a (unimportant) zwitterionic as well as a (very important) bi-radical boundary structure in S and O. The latter convincingly explains two primary reactions. The latter convincingly explains two primary reactions of dimethyl sulfoxide with hydroxyl radicals:

1. If DMSO binds a hydroxyl radical via *oxygen*, the *hydro-peroxyl* radical is separated in addition to volatile dimethyl sulphide. From this, oxygen •O-O• can be split off from origin with another •OH-radical, forming H₂O. Excess volatile, unpleasant smelling and toxic dimethyl sulphide (DMS) is excreted mainly through the lungs ²⁵. The functional group is therefore **O=S**.
2. If DMSO binds a hydroxyl radical to the *sulphur (S=O)*, the following HO• attacks the *dimethyl sulfoxide* by oxidation to kidney-permeable dimethyl sulfone (DMSO₂ ²⁵) after splitting off water. DMSO *must* be reckoned with the development of nascent oxygen or via the formation of dimethyl sulfone to the group of direct radical scavengers. DMSO₂ is excreted in the urine. - Its antioxidant metabolism has also been demonstrated by toxicological and clinical studies ^{25,31-33}. Properties of this super-solvent and "tractor molecule" for (*conceived as geometrically flat!*) DMSO are probably based on bi-radicality in combination with *out-off-plane* oscillations.

* Note: A *direct* radical scavenger disposes of reactive oxygen radicals via functional groups and not via enzymes. This means, that the interception and "neutralisation" of radicals is extremely fast ³⁰.

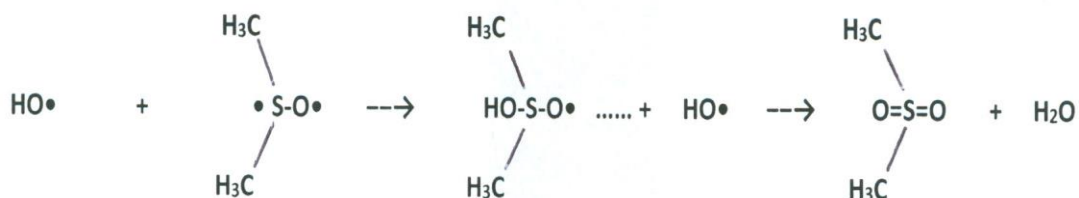
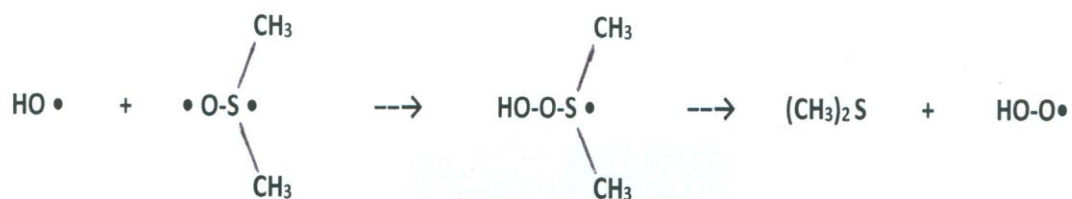


Fig. ② Probable degradation modes of DMSO e.g. by hydroxyl radicals

If there are also multiple bonds in molecules (apart from the functional groups mentioned for the conversion of the radicals) or possibly bonds preformed for rearrangement, then slower radical reactions are also possible. They should *also* be detected by the ab initio methods ^{*,30}. In the present article, however, only reactions with ultrafast kinetics involving reactive species are of interest as examples.

* Note: Therefore, quantum pharmacological methods for testing multiple reaction sequences in "radical diseases" could also contribute to cost reduction in the health care system ³⁰.

3. Selenous acid oxidises hydroxyl radicals - a consuming reaction

Selenous acid (H_2SeO_3) is a component of easily soluble sodium selenite pentahydrate in aqueous physiological milieu and primarily acts directly as a radical scavenger: hydroxyl radicals are oxidised to hydrogen peroxide, whereby selenic acid consumes itself in a reductive degradation ^{34,18}. The nascent selenium produced in this process is excreted renally after glycosylation and glucuronidation in the form of selenium sugars and *trimethylselenoniumchloride* and/or exhaled after methylation. Mono- and di-methylation are already regarded as signs of an incipient but reversible selenium intoxication ³⁵⁻³⁷. What is of great physiological importance, however, is that H_2O_2 has a half-life up to 10,000 times longer than hydroxyl radicals and can, for example, be disposed of in the metabolism of the thyroid by HI both directly and enzymatically ^{18,38}. Therefore, H_2O_2 is far less aggressive in tissue than $\text{HO}\cdot$ -radicals. Selenous acid has a direct (i.e. per se and without enzymes) anti-inflammatory effect in tissues ^{34,18,30}.

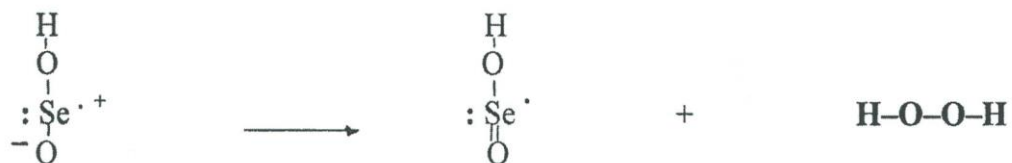
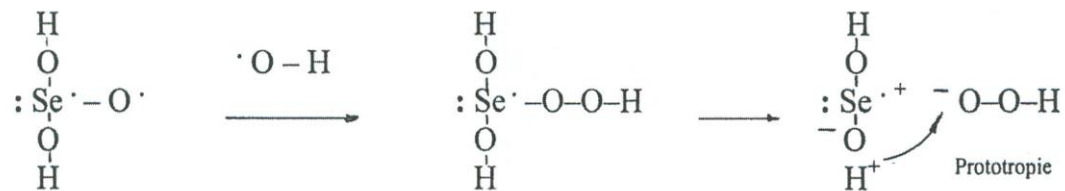
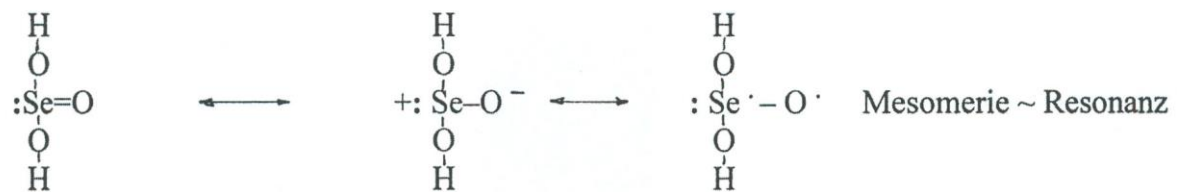


Fig. ③ Hydrogen peroxide generation by selenous acid from sodium selenite ³⁴

For years, the methods of theoretical physics integrated into computational chemistry have been helpful in attempts to describe an assumed reaction process. Why? - In the post-Schrödinger era, so-called "ab initio methods" were developed for calculating chemical binding energies and associated vibrational states as well as possible models of molecular structure resulting from them. These are based solely on internationally recognised atomic parameters, natural constants, the time and the Born-Oppenheimer approximation³⁹ for molecular ground states. By means of such methods, molecular structures involved in numerous reactions (close to the ground state) can be theoretically represented in advance in good agreement with experimental (e.g. vibrational spectroscopic) analyses. -

If chemists postulate hypothetical intermediate stages (for such - near ground state - reactions) on the basis of empirically obtained ideas using all possible boundary structures, these can be compared continuously and up to the end of the reaction with the quantum chemical results mentioned. If the predicted structural assumptions of the intermediates are coherent with the theoretical calculations, the chemical mechanism is probably conclusive and therefore prognostically justified. Accordingly, the disposal of HO• - radicals with the reductively consuming selenic acid could be graphically represented by ab initio calculations (coupled with density functional methods) in the computer-associated design^{30,32,34}. This procedure is probably also true for processes (*partial radical-controlled oxidative and metabolic*) in the opposite direction of the *reabsorption of selenium*. They were not taken into account in earlier investigations. Coherence testing procedures should therefore continue to accompany our thinking in steady-state equilibria in the future.

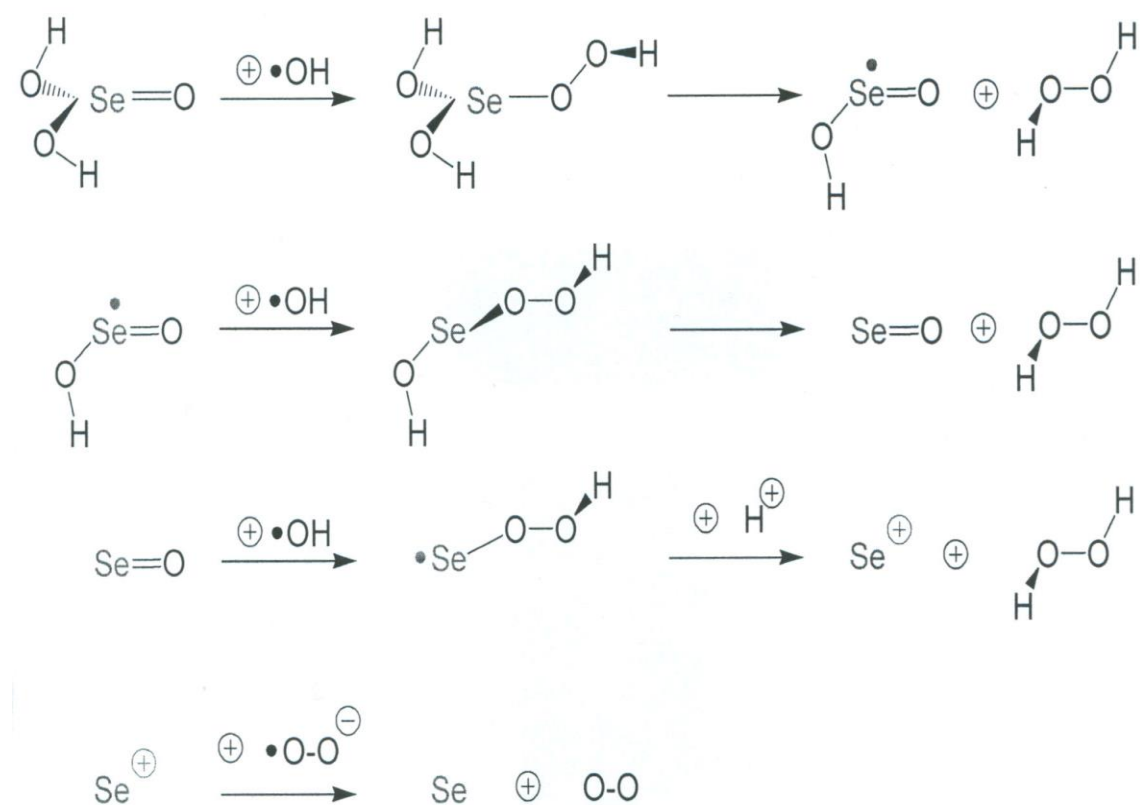


Fig. ④ Consumptive disposal of hydroxyl radicals by selenic acid according to quantum pharmacological calculations^{30,34}

4. Dexamethasone dimerizes hydroxyl radicals - a catalytic reaction

From the well-founded assertion that selenic acid is primarily a direct radical scavenger and thus ad hoc anti-inflammatory, it is necessary to compare it with known corticoids that also have a direct anti-inflammatory effect.

The choice fell on dexamethasone as the pharmacological lead substance of the synthetic cortisone derivatives³⁴. Molecular structures show that natural hormones of the adrenal cortex such as cortisol and cortisone, but especially the stronger anti-inflammatory synthetic derivatives (e.g. methylprednisolone, betamethasone, budesonide^{*})¹ have functional groups comparable to dexamethasone: There are also the polar carbonyl components with up to two adjacent CC - double bonds in the A - rings. These functional groups catalytically dispose of hydroxyl radicals by dimerising. Other properties of glucocorticoids will not be discussed in the context of the present topic. - According to the quantum-chemical calculations mentioned above, the disposal of hydroxyl radicals by dexamethasone (1) proceeds via the following catalytic cycle: The attack of an $\bullet\text{O-H}$ -radical on the carbonyl group of dexamethasone results in a very unstable peroxy-dexamethasone derivative (2), which is radicalised with $\text{H-O-O-C}\bullet$. This immediately binds another $\bullet\text{O-H}$ - radical. This immediately binds another $\bullet\text{O-H}$ - particle at the C \bullet - locus. Hydrogen peroxide (H-O-O-H) is now spontaneously split off from the extremely unstable peroxy-hydroxy structure (3) and thus the stable dexamethasone molecule (1) is recovered^{34,30}.

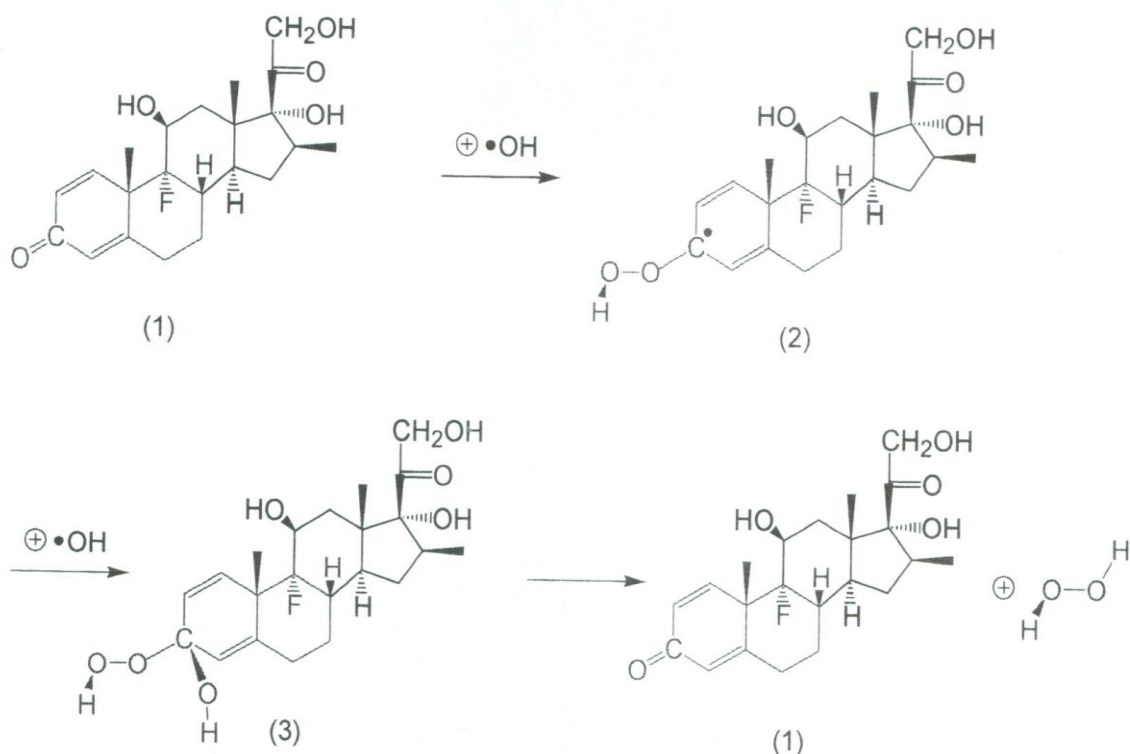


Fig. ⑤ Catalytic disposal of hydroxyl radicals by dexamethasone according to quantum pharmacological calculations^{30,34}

* Note: Budesonide is a synthetic corticosteroid that can also be inhaled as a powder (cf. fig. 18).

5. iso-energetic reactions and valence isomerism

Even ab initio and density functional methods are sometimes not free of supposed uncertainties. It is possible, for example, that a high-performance computer discovers energetic equivalent alternatives while checking predicted reactions. In such a case, two different reaction paths could be taken.

5.1 Selenous acid

However, it is also possible to reach a "kinetic dead end" (with the consequence of a virtual "chemo brake")^{34,38}. However, the reaction path leads out of the aforementioned dead end back into (more familiar) passive redox patterns while maintaining the $\bullet\text{OH}$ -reactions. A kinetic vicious circle is impossible under these conditions.

5.2 Dexamethasone

In contrast, a chemically real "circle" in the steady state subsequently signals a different reduction-oxidation strategy. It presents itself in two auto-catalytic processes. Both reaction pathways are to be demonstrated with the $\bullet\text{OH}$ radical disposal by dexamethasone³⁴. In the case of dexamethasone, the quantum chemical calculation also results in an iso-energetic evolution of hydrogen peroxide via hydrogen bonds.

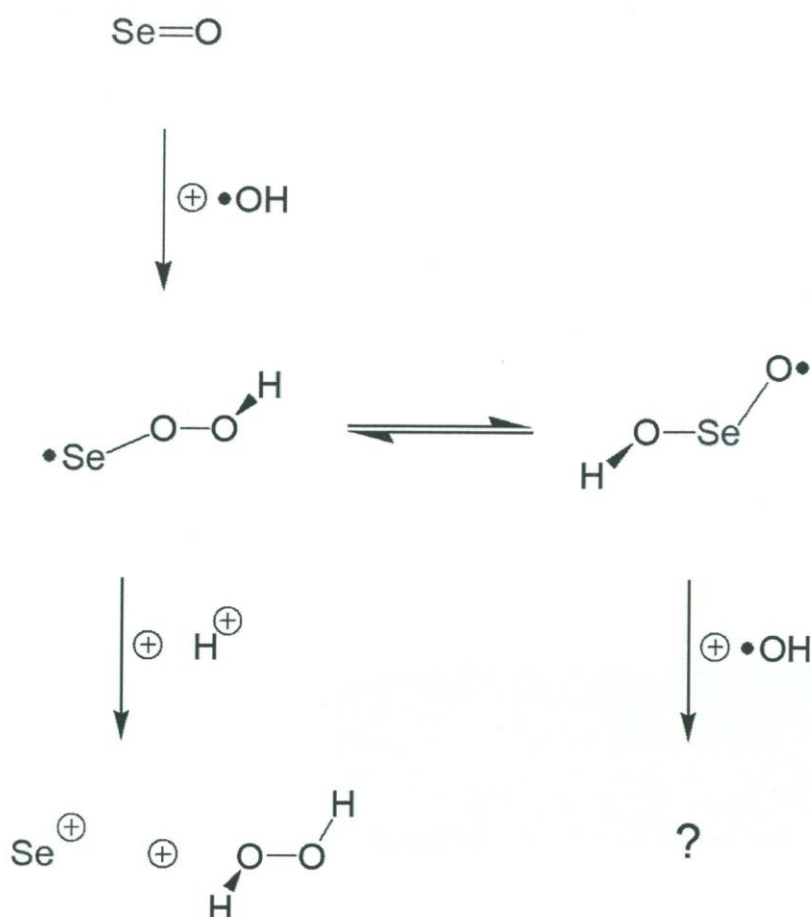


Fig. ⑥ Selenous acid: $\bullet\text{OH}$ -disposal - here as a "kinetic dead end"^{30,34}

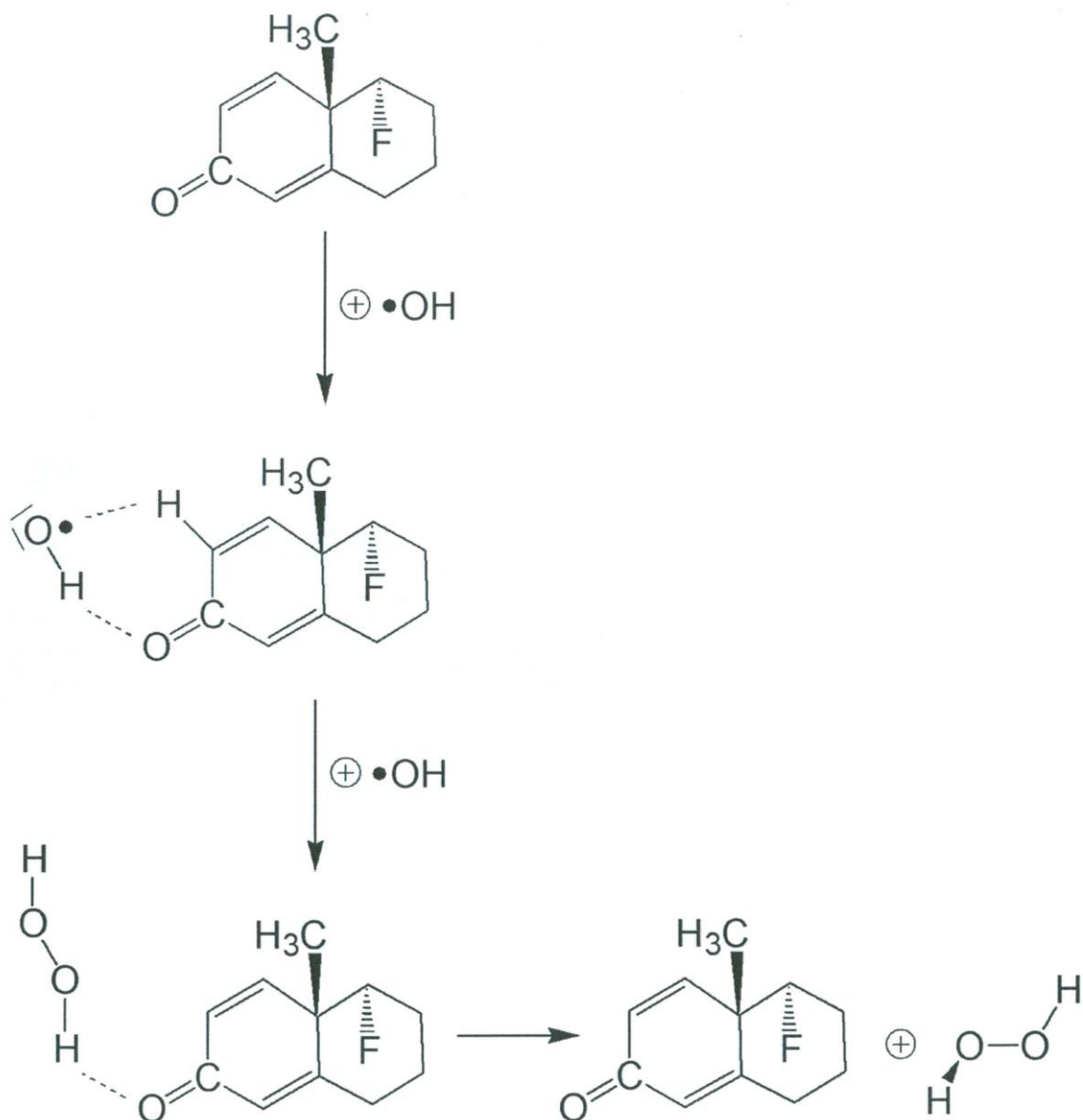


Fig. ⑦ Dexamethasone: autocatalytic $\bullet\text{OH}$ -disposal via hydrogen bonds ^{30,34}

5.3 Reabsorption of selenium

Under deficiency conditions, selenium is "recycled" : i.e. selenium sugars and/or methylated selenium molecules are not immediately excreted renally. In addition to the highly toxic hydrogen selenide (SeH_2), they are radically oxidised back and fed into the metabolism. In an acidic medium, superoxide anion radicals and hydroxyl radicals (from HOOH) would be suitable for this. Only by way of reabsorption do metabolites of dietary selenium or from food supplements, e.g. selenium-methionine, also indirectly acquire the properties of direct radical scavengers, even if the oxidation states +6 of selenate or +4 of selenite are not reached during reabsorption. - Physiological reabsorption mechanisms have the effect that trace elements are more difficult to detect and often cannot be correctly weighted in their importance.

6. Oxygen acids (oxoacids) and hydroxyl radicals

Do oxygen acids have antioxidant-relevant functional groups compared to •OH radicals? This question should be tested using known examples of simple acids (6.1-6.4). In the assumed mechanisms, functional groups $X=O$ of these acids are attacked by at least one •OH radical, whereby H_2O_2 is (or could be) formed. H_2O_2 - evolution means oxidation for hydroxyl radicals and reduction of functional groups of oxo acids. On the other hand, there are functional groups of oxoacids that are not reduced by radicals (6.5). The examples of oxygen acids were taken from WIKIPEDIA.

The examples show: Oxoacids contain oxygen by definition. However, they do not necessarily have an oxidative effect. Each oxygen double bond generates mesomeric radical structures. From this, bactericidal and virucidal but also per se anti-inflammatory effects become understandable. However, hydroxyl radicals never react with the HO-P bonds of phosphoric acid in the organism to form H_2O_2 !

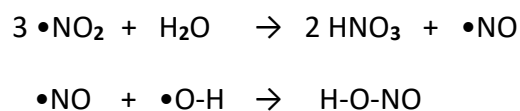
6.1 Nitric acid, nitrogen oxides ⁴⁰ and •OH-radicals



Fig. ⑧ Nitric acid: reduction to nitrogen dioxide

Nitric acid, nitrous acid and nitrogen oxides in the lungs according to BfR ⁴¹:

In accidents involving concentrated nitric acid, bronchi can be exposed to high concentrations of •NO₂. The uptake of •NO₂ in the respiratory tract ultimately results in HNO₃ and HNO₂ under local inflammation with the involvement of •NO. •NO₂ disproportionated in the aqueous solution of the alveoli to nitric acid and nitrogen monoxide:



Nitrogen oxides •NO₂ and •NO per se lead to inflammatory irritations in the alveoli. Nitric oxide reacts with •OH in oxidative stress (inflammation) to form nitrous acid. As these reactions progress, oedema may develop in the lungs.

In medicine, esters of nitric acid serve as •NO donors. They are prescribed in angina-pectoris crises ¹ and •NO is then supposedly enzymatically released from the corresponding pharmaceuticals. H_2O_2 is also disposed of enzymatically.

Some lower organisms such as bacteria, fungi and plants require *enzymatically* acting nitrate reductases to catalyse the reduction of nitrate to nitrite in the acidic micro-environment ($+H^+$) ⁴².

6.2 Chloric acid, chlorine oxides⁴³ and •OH - radicals

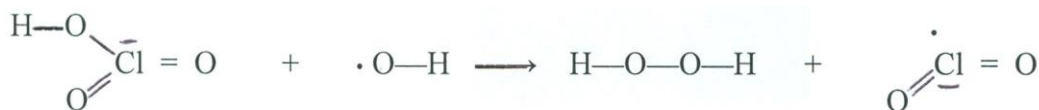


Fig. (9) Chloric acid → chlorine dioxide

Remark:

Chloric acid is a strong oxidising agent. Highly diluted, aqueous potassium-chlorate solutions used to be used to gargle in the mouth and throat area for inflammation (acidic microenvironment)^{1,44}.

6.3 Sulphuric acid⁴⁵, sulphur oxides and radical boundary structures

Remark:

Reactions of the acids H₂SO₄ and H₂SO₃ and their anhydrides SO₃ and SO₂ are often carried out exclusively by means of ionic boundary structures. This does not result in an adequate understanding of the respective reaction.

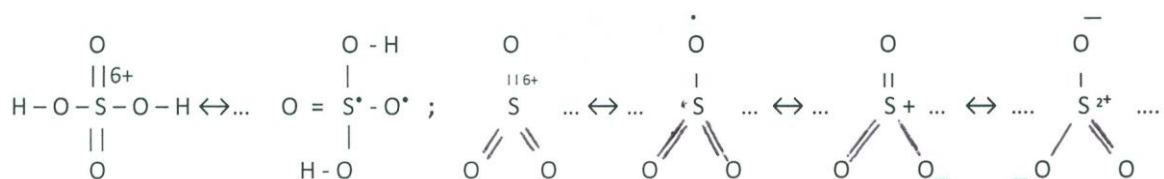


Fig. (10) Radical and ionic boundary structures of H₂SO₄ and the anhydride SO₃



Fig. (11) Radical and ionic boundary structures of sulphurous acid and sulphur dioxide

Remark:

In addition to ionic structures, it is essential to consider mixed-ionic-radical and pure-radical boundary formulae to understand a kinetic snapshot.

Non-ionic compounds result from homo-syntheses and/or homolyses. They can only be understood in this way. It therefore seems advisable to consider from the outset also, if possible, such boundary formulae that correspond to radical-induced processes. Therefore, besides ionic also radical and mixed boundary structures have to be considered. For example, the reaction of a hydroxyl radical with sulphuric acid (not highly concentrated, not hot) results in a "hydro-peroxy-sulphuric acid". Further comparable $\bullet\text{OH}$ reactions and H_2O_2 splitting also lead to the volatile anhydride SO_2 via H_2SO_3 . A theoretical "preliminary test reaction" with $\bullet\text{OH}$ -radicals must make experimental-chemical processes appear at least probable or acceptable. In aqueous, diluted H_2SO_4 solution, at normal temperature and pressure (NTP), ionic reactions thus dominate. - The comproportionation of identical-radical molecules results (depending on concentration and temperature) in the rarer and more unstable compounds the dithionic acid ($\text{H}_2\text{S}_2\text{O}_6$) and the dithionous acid ($\text{H}_2\text{S}_2\text{O}_4$). Attacks of $\bullet\text{OH}$ on the respective acid functions also result in the release of SO_2 after successive cleavage of H_2O_2 . - The highly concentrated Sulphuric acid (Oleum) has an oxidative effect with increasing temperature. With concentration and increasing temperature, various radical processes then begin to replace ion reduction-oxidation kinetics in mixed mechanisms. The redox reactions no longer follow an exclusively ion-theoretical interpretation ⁴⁶.

A possible reaction mode for hot, concentrated sulphuric acid with a pure copper surface is now to be proposed, whereby this proposal would still have to be checked for its quantum physical coherence. Under the given conditions, the following assumptions form the starting point: SO_3 can radically attack both the copper surface and water molecules. Starting from a bi-radical SO_3 - boundary structure, perhaps $\text{CuO}\bullet$ and SO_2 are formed from copper, because copper reduces SO_3 . SO_2 is volatile. The SO_2 remaining in the system is oxidised by further $\text{HO}\bullet$ - radicals to Sulphuric acid, or to SO_3 and water. If, on the other hand, the bi-radical SO_3 -structure splits water, H_2SO_4 or SO_3 and H_2O are formed.

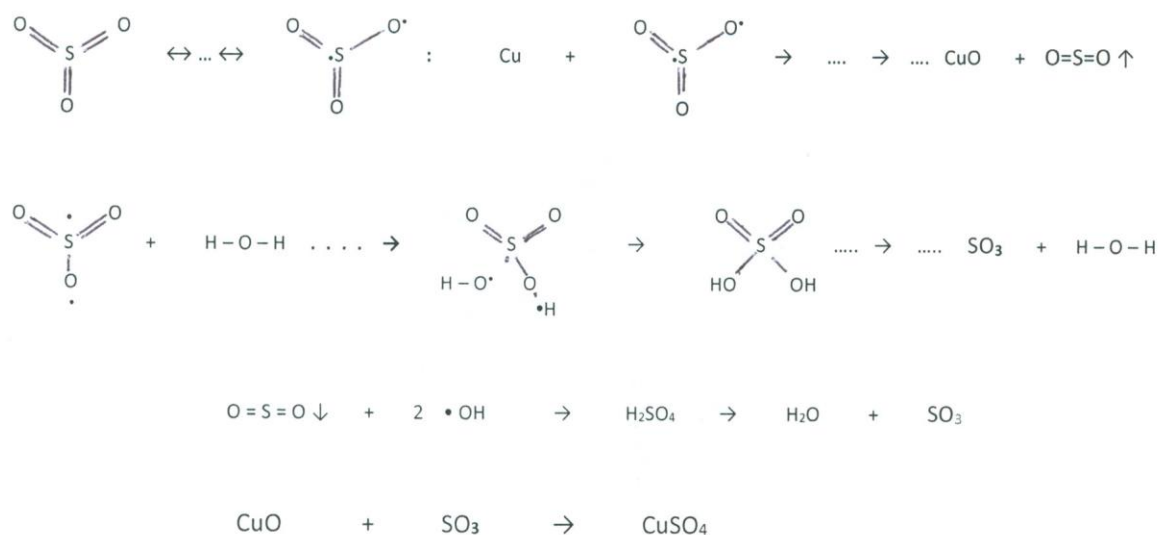


Fig. ⑫ Assumed reaction: concentrated, hot sulphuric acid with copper

Presumably, a more complicated mechanism is at work: Copper (Cu) perhaps statistically coordinates the HO-groups of the sulphuric acid at its lattice boundary, then catalytically splits off •OH to form nascent CuO• radicals, which immediately reacts with SO₃.

•OH-radicals are furthermore surface-active themselves. No atomic oxygen is formed. Free H⁺, HO¹⁻ - and Cu²⁺ - ions are most likely not formed. - Possibly there is a complex "ion-radical mixing mechanism".

Under physiological conditions, sulphuric acid - as shown - is not an oxidising agent and is therefore not suitable as a "radical scavenger".

6.4 Permanganic acid (Manganese-VII acid), potassium permanganate ⁴⁷ and •OH-radicals:

Hydroxyl radicals lead to the reduction of permanganic acid in an acidic aqueous medium with the splitting off of hydrogen peroxide up to the Mn³⁺ stage with 4 valence electrons left.

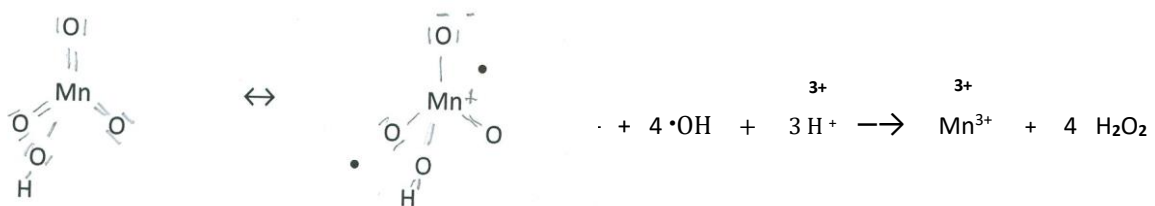


Fig. ⑬ Partial reduction of permanganic acid by hydroxyl radicals

* Notes on permanganic acid and permanganate ^{47,48}:

Permanganic acid ⁴⁷ is one of the strongest and (therefore) most unstable oxidants. The course of oxidation is largely dependent on the pH value of the chemical environment. HMnO₄ probably only exists as a completely dissociated acid in aqueous solution. This would result in the following decay patterns:

1. A partial decomposition of the free acid alone under radical formation.
2. Degradation under the influence of radicals and reducing agents
3. Reactions by reducing agents alone

Permanganate in acidic solution: Reducing agents additionally achieve degradation to Mn²⁺. Potassium-permanganate is often used as an oxidising agent.

Potassium-permanganate in medicine ⁴⁸:

The disinfecting, virucidal KMnO₄-solutions used to be used - like potassium-chlorate solutions (see above) - as gargles for inflammations of the mouth and throat and, among other things, for rinsing mucous membranes and wounds as well as for gastric lavage e.g. after food poisoning by enterococcus toxins.

Furthermore, drinking water was disinfected. Radical-based disinfection takes place predominantly in an acidic environment. Permanganate-solutions can be used to eliminate algae in stagnant waters. KMnO₄ -solutions also served as deodorants.

6.5 Phosphoric-acid

Note: This acid does not split off H₂O₂. (See also section 6 beginning.)

7. Ammonia, nitrogen oxides, nitrates and nitrites - associated problems

7.1 Biologically relevant molecules and ions in the nitrogen cycle ⁴⁹

Nitrogen exists in the oxidation states from -3 to +5, partly naturally but mainly in synthesis products. Besides gaseous NH₃, N₂, N₂O, •NO and •NO₂, anions of some oxo-acids and organic •NO-compounds are important for the living world.

In the context of disaster medicine, the following molecules of nitrogen are considered ecologically relevant: Ammonia (NH₃) and nitrous oxide or "laughing gas" (N₂O). They originate predominantly from agriculture (with factory farming). Emissions of nitrogen-monoxide (•NO) and nitrogen dioxide (•NO₂) are mainly attributed to industry, households and traffic (as pollutants). The highly oxidising dinitrogen-tetroxide (N₂O₄), which has been used as rocket fuel among other things, cannot yet be conclusively classified.

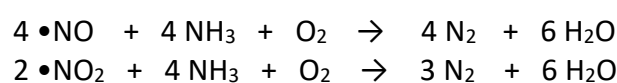
Of the oxoacids of nitrogen, nitrates (NO₃⁻), nitrites (NO₂⁻) and peroxy-nitrites (ONO₂⁻) are also biologically and medically significant. - Nitrate ions from fertilisers are not considered toxic in low concentrations. However, they also endanger human health in the long term if soils are overfertilized.

Organic nitrates and nitrites are indispensable components of cardiovascular drugs as "NO-donors" (endothelium-derived relaxing factor or EDRF). When incorporated in excess, nitrites have a predominantly haemato-toxic effect and are also carcinogenic through nitrosamine formation. They are added by law (strictly regulated) to the "curing salt" in the production of sausages to kill life-threatening botulism bacteria. Nitrites are also used (sometimes illegally) to preserve the "fresh" colour of meat.

Peroxy-nitrite (ONO₂)⁻ can be formed in the tissue from •NO and superoxide anion radicals (•O₂⁻). Similarly, hyponitrous acid (N₂(OH)₂ ⇌ HONHNO) is formed during inflammation in a weakly acidic environment. Highly reactive, toxic peroxy-nitrite is used - in an immunity-stabilising manner - by cytotoxic cells to kill pathogens.

7.2 Nitrogen oxides from combustion processes and their reduction ^{50,51}

•NO and •NO₂ in short •NO_x are components of combustion gases from fossil energy sources. These come mainly from coal and oil from land vehicles, air traffic, shipping, heating, households, industry and waste incineration plants. The main share of •NO_x emissions in traffic comes from diesel-engines. Nitrogen monoxide is the primary emission. Natural sources of nitrogen oxides are lightning discharges in thunderstorms. •NO and •NO₂ act, among other things, through acid formation. They irritate mucous membranes and are toxic. - The reaction of •NO and •NO₂ with ammonia (NH₃) and oxygen has proven to be a successful large-scale selective non-catalytic reduction of nitrogen oxides (SNCR):



•NO_x is naturally degraded in the atmosphere in numerous complicated radical reactions. This occurs near the ground in hours but in the upper troposphere over days. However, the fact that ammonia from agriculture contributes significantly to the reduction of air quality must not be overlooked.

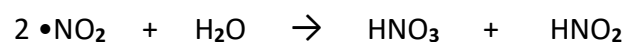
7.3 The Diesel exhaust debate ^{52,53}

It was pushed and commented on by a group of lung specialists, who are close to the car industry. However, there is an international consensus among pulmonologists regarding the toxicity of nitrogen oxides, especially •NO₂ from diesel exhaust. The short-lived radical •NO is rapidly oxidised by atmospheric oxygen. •NO₂ is considered to trigger severe respiratory distress, bronchitis, coughing fits and asthma when exposed to high levels during the day - especially for people with a medical history. With prolonged exposure, nitrogen oxides are considered a source of chronic respiratory diseases with constriction of the bronchial tubes and their blood vessels, as well as subsequent cardiovascular problems with a lower life expectancy. Diabetes is considered a frequent side effect of constant exposure to nitrogen oxides ⁵⁴ - The following European limit values ⁵⁵ have therefore been set for nitrogen dioxide:

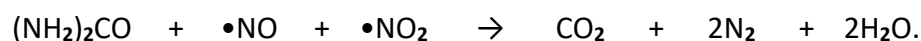
Annual mean value as •NO₂ - Long-term limit value: 40 µg / m³ air
One-hour mean value as •NO₂ - Short-term limit value: 200 µg / m³ air

Note: The short-term limit value may not be exceeded more than 18 times within one year at the same measuring point. (Cf. also Official Journal of the European Union of 29. 06. 2007 ^{50,52,53}).

Nitrogen dioxide, like CO₂ and SO₂, is a source of acid rain:



In order to reduce the nitrogen oxide content in diesel exhaust gases, the "AdBlue® process" has proven its worth ⁵⁶. In this process, a 32.5% urea aqua-destillata solution is sprayed in doses into the exhaust passage between the diesel particulate filter and the selective catalytic converter (SCR). This reduces •NO and •NO₂ to N₂ and water vapour by means of NH₃. - Apart from the engine type and the driving style of the diesel driver, material science, thermodynamic and technical problems (installation space, AdBlue tank size, etc.) limit the reduction depending on the vehicle type in accordance with



Driving style and technical problems could reduce the desired exhaust gas detoxification ⁵⁷. Even before the polemic-political diesel exhaust debate, the realisation of nitrogen oxide detoxification was known to be an urgent task. The AdBlue process has actually calmed down the discussion somewhat.

7.4 Nitrogen oxides and pollutants from aircraft

Thanks to a Swiss initiative, aircraft have recently been subjected to emissions tests. In the process, the combustion of kerosene in jet engines provides as a material balance per flight hour (e.g. for a commercial aircraft with 150 seats) ⁵⁸:

Air sucked in: 850.000 kg - fuel kerosene: 2700 kg - emitted cold air: 722.700kg.
 Exhaust gases and hot air: 130.000 kg. Thereby, the exhaust gases from jet engines with a weight of approx. 11.835 kg as greenhouse gases 8.500 kg CO₂ and 3.300 kg H₂O i.e. water vapour as well as pollutants: 30 kg •NO_x + 2.5 kg SO₂ + 2.0 kg CO + 0.4 kg HC + 0.1 kg particulate matter. The exhaust gases are taxed with additional levies at the airport.

The Federal Ministry for the Environment, Nature Conservation and Nuclear Safety, together with partners from 16 countries, industries, research institutes and universities, is funding the < Clean Sky > project to improve efficiency potentials by incorporating modern engine technology and energy-saving developments ^{59,60}.

Recent studies on the influence of nitrogen oxide emissions in contrails on the composition of the upper troposphere and lower stratosphere measured in the wake of aircraft show that of all the reactive nitrogen compounds NO_y (•NO, •NO₂, HONO, HNO₃, N₂O₅.), between 80% and 95% •NO is produced at the engine outlet (in addition to HO• and H₂O as well as CO, C and SO₂, among others). Nitrous acid (HONO) is the most important component and reservoir gas for •NO- and HO•- radicals. It condenses with water vapour on catalytically acting soot particles (M) visibly as condensation trails to form temporary ice crystals ⁶¹.

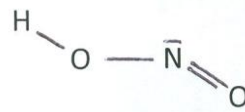
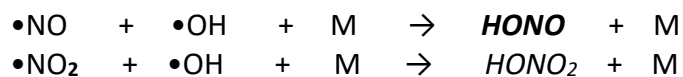


Fig. ⑭ Nitrous acid in trans-planar configuration

With a high number of possible questions, the exact determinations depend on many parameters per aircraft type. These could be calculated in individual cases using the "atmosfair-CO₂ emissions calculator" ⁶². The above examples basically prove the considerable emission of pollutants with increasing flight operations, regardless of considerable amounts of climate-damaging components such as CO₂ and water vapour. Moreover, due to aircraft noise, a farewell to kerosene ⁶³ would be desirable.

7.5 Nitrogen oxides and pollutants from shipping ^{64,65}.

Container ships are indispensable to the global economy, as about 90% of trade is seaborne. But like container giants, almost all luxury cruise liners run on heavy fuel oil, which is harmful to health and the environment. This is the last viscous fraction from the processing of petroleum into kerosene, petrol and diesel.

In June 2019, the environmental organisation T & E <European Federation for Transport & Environment> published a critical study on air pollution by cruise ships entitled "One Corporation to Pollute Them All - Luxury cruise air emissions in Europe" ⁶⁶. According to this study, 203 cruise ships of the world's second largest corporation <Royal Caribbean Cruises> emitted approx. 15% of the nitrogen oxides (corresponding to 155kt •NO_x) of all passenger cars registered in Europe in 2017. SO₂ emissions amounted to approx. 62kt. Particulate matter with high soot content was approx. 10kt. - Port cities like Marseille are particularly polluted: in 2017, 57 cruise ships docking here produced 15.219 kg of SO₂, about 4 times more than 339.987 registered passenger cars (4144 kg SO₂).

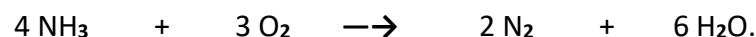
Port cities in Mediterranean countries are most affected by sulphur dioxide pollution, followed by other ports on non-Mediterranean coasts in Europe. All coastal regions of Spain are very strongly affected. In the case of sulphur oxide emissions from sulphate aerosols, countries located further north are also affected. For example, along the sulphur emission controlled coastal region of Denmark in 2017, SO₂ pollution has been noted equivalent to 18 times the SO₂ emissions of the approximately 2.5 million passenger cars registered in Europe that year ⁶⁶.

Nitrogen oxides and sulphur oxides directly endanger the health of people living near the coast in port cities due to docking containers and luxury liners.

As an exceedingly harmful gas, SO₂ also brings acidification of water and soils far inland. Carnival Corporation cruise brands dominate SO₂ air pollution ⁶⁶: 47 of their cruise giants emit 10 times more SO₂ than 260 million passenger cars in Europe. Second place, with 4 times more emissions, is held by the Global Royal Caribbean Cruises-Company, as noted above.

The EU-Commission was unable to reach a decision on solid pollutant limits at the International Maritime Organisation (IMO) and therefore could not push through the necessary stricter environmental guidelines against the interests of the World Shipping Council. This body represents shipping companies that primarily earn money from liner traffic with container ships.

Because of the "heavy oil emissions scandal", alternative propulsion components (batteries, marine diesel, liquefied gas?) are also being planned. The Norwegian state-owned energy company *Equinor* ⁶⁷ is already experimenting with ammonia (NH₃) as perhaps the only and in future most environmentally friendly fuel for ocean-going vessels according to



However, the following problems arise: Battery propulsion is only possible for short distances. A "marine diesel scandal" would not be a solution. Liquefied gas poses considerable risks in the event of an accident (for immediate safety and for the environment through methane) ⁶⁷.

7.6 Nitrate in drinking water ⁶⁸.

The pollution of groundwater and thus also of drinking water with nitrogen inputs from slurry, manure and artificial fertiliser from agriculture has been known for many years. Excess nitrogen compounds that are no longer absorbed by plants are released into the atmosphere (e.g. N_2O) or, after oxidation, are washed out with the rain as nitrates and are introduced via surface waters into the groundwater and thus into the drinking water supply. 50 mg nitrate per litre of drinking water was set as the upper limit for drinking water, which is in itself well monitored, in accordance with the European Nitrate Directive 91/676 EEC. This value was exceeded again and again at numerous measuring points in Germany *after* 1991 - even *after* the Fertiliser Ordinance was *only* passed in 1996 ^{68,69}. Nitrate is a danger, especially for infants with increased bacterial conversion to nitrite and still insufficient production of gastric acid. Exposure can develop into methaemoglobinaemia (cyanosis) ⁶⁹. A Danish study states that the European guideline value of a maximum of 50 mg nitrate per per litre of drinking water is considerably too high in the case of perennial uptake, and already clearly significantly lower values correlate with a significant risk of colorectal cancer ^{70,71}.

The removal of nitrate from drinking water is associated with complex and expensive processes of reverse osmosis, ion exchange, electrodialysis or biological denitrification by microorganisms. Therefore, the BUND study ⁷² sums up that without a fundamental legal and controlled reduction of livestock density with an ecologically compatible fertiliser law, normal consumers will continue to finance agriculture through taxes and food costs.

8. Anthropogenic contributions to global warming ⁷³

8.1 Natural greenhouse gases ⁷⁴

The natural greenhouse effect ⁷⁴:

Molecules, molecular associations and passive solids (molecules), which absorb infrared radiation from the earth's surface and partially reflect it, cause a warming of the troposphere in energetic dynamic equilibrium with the spectrum of the irradiated sunlight. These particles are considered "natural" greenhouse gases. In the history of the earth, they caused a temperature rise of $-18\text{ }^\circ\text{C}$ to $+15\text{ }^\circ\text{C}$ at which life was supposedly able to develop. These still include the "trace gases" carbon dioxide (CO_2), methane (CH_4), nitrous oxide (N_2O) from agriculture and ozone (O_3) produced near the ground. - An extremely effective greenhouse gas is water vapour with condensates in the form of ice crystals in cloud formation and precipitation, which cooperate in many ways via hydrogen bonds. Natural aerosols must also be included. - Temporary solids also include soot particles and sulphur compounds from volcanic eruptions as well as dusts of various origins (e.g. Sahara dust). The above molecules and particles largely contribute to global warming independently of humans.

8.2 Key messages of the IPCC Fifth Assessment Report - Climate Change 2014

Synthesis Report ⁷⁵

Observed climate change:

With strong annual fluctuations due to weather conditions, the average daily mean temperature in Germany rose by about 0.11°C per decade between 1880 and 2020, measured in spring and autumn. The warming in the five decades between 1970 and 2020 was significantly faster, at around 0.18°C. Apart from regional extreme precipitation, fewer cold and more hot weather periods were recorded. With decreasing snowfall in winter and permanently dramatic glacier melting, a rise in sea level of around 20 cm has been recorded since 1900.

8.3 Anthropogenic greenhouse gases ^{76,77}

The additional greenhouse effect:

With economic growth, steadily increasing prosperity and a rapidly growing population density was accompanied, especially in the northern hemisphere, by previously unobserved emissions of the infrared-active greenhouse gases carbon dioxide, methane and nitrous oxide. - The energy introduced into the environment causes a strong heating of the troposphere. It is still stored predominantly (approx. 90%) in the warming oceans and can thus hardly be perceived by the individual. ⁷⁸ - The masses of plastic waste disposed of in the oceans are also degraded to carbon dioxide, albeit extremely slowly. - Due to the enormous CO₂ input, lakes and seas are becoming more acidic, damaging marine ecosystems (e.g. coral reefs) ⁷⁹. With falling pH values, bacteria and viruses will proliferate in waters. - A continuously rising temperature will correlate with crop failures, especially for people in poorer countries (with increasing plagues of locusts), and will also affect the last resources of food and drinking water. Tropical insects will also become native to European countries with the northward shift of Mediterranean vegetation and thus import dangerous pathogens. If it is not possible in the foreseeable future to limit global warming to below 2°C by drastically reducing greenhouse gas emissions, sea levels will rise with irreversible damage (up to the metre range). The consequences of global droughts, devastating storms, widespread flooding of many coasts, dam failures and more frequent weather extremes will probably be compounded by unimagined social problems due to large-scale migrations from severely affected regions. Distribution struggles to secure livelihoods, especially food and drinking water, can be expected and will have a lasting impact on the northern hemisphere.

Reduction targets and adaptation options ⁸⁰: In its Fifth Assessment Report, the Intergovernmental Panel on Climate Change (IPCC) attempts to present various scenarios (RCPs) for "interested parties" - not least for politicians as "decision-makers" - under which compliance with the 2°C upper limit (?) still appears to be feasible ⁸¹. To reduce greenhouse gas emissions, new key technologies are needed that can guarantee a measurable (not further negotiable) increase in carbon-free or low-carbon energy supply. Will hydrogen (H₂) as an energy source perhaps solve all the problems of future energy demand? - If the use of fossil fuels, including bioenergy, continues to be regarded as "indispensable", it will be necessary to couple carbon dioxide capture with CO₂ storage. - Irrespective of the special problem of demand, the Intergovernmental Panel on Climate Change considers a temporary continuation of nuclear energy to be indispensable until 2050 in order to achieve the 2°C

target. It is incumbent on us "affluent citizens" to familiarise ourselves with unavoidable (hopefully transparent!) and justified cost increases of politically ambitious measures. This includes changes in the outmoded behaviours of a consumption-driven lifestyle - i.e. a "rethinking for the purpose of redirection" in ourselves.

8.4 Svante Arrhenius and the global warming (1896) - The Ion Theory of Electrolytic Dissociation ^{83,84} - Friedrich Wilhelm Ostwald: Catalysis (1909) ^{85,86}

In his dissertation in 1884, S. Arrhenius formulated "The Ion Theory of Electrolytic Dissociation". It expanded the views of chemists, which was also recognised with the Nobel Prize in 1903. Less well known is his discovery of CO₂ as a greenhouse gas in 1896 ^{83,84}. According to his own extensive calculations, the visionary researcher predicted a "beneficial" warming of the troposphere by four to six degrees if the proportion of carbon dioxide in the atmosphere doubled. The ion theory contributed nothing to the understanding of the radical chemical basis of global warming.

Arrhenius' patron, a renowned electrochemist at the time, was Friedrich Wilhelm Ostwald. According to his "dilution law" ⁸⁵ the dissociation of an electrolyte depends on the initial concentration of the electrolyte and its degree of dissociation. This tends towards one with increasing dilution. F. W. Ostwald was - without knowing it himself - an excellent radical chemist: the synthesis of ammonia by reducing nitrogen from the air goes back to him. It became known as the "Haber-Bosch Process". He also founded the large-scale synthesis of nitric acid by oxidising ammonia (known as the "Ostwald Process"). In 1909, Ostwald was awarded the Nobel Prize in recognition of his work on catalytic processes in inorganic and organic chemistry ⁸⁶. His definition of a catalyst is still valid today. However, Ostwald was little interested in greenhouse gases and the problems of global warming.

8.5 Walther Kossel and his Valence theory of the chemical bond (1916) ^{87,88}

Chemistry owes final clarity about the existence and nature of ions to the physicist W. Kossel, a student of Philipp Lenard and Arnold Sommerfeld. He last taught as a full professor of theoretical physics at the Eberhard Karls University Tübingen. - According to X-ray structure analysis, salts exist in crystals (independent of solvents) as cations and anions. The structure of salts becomes understandable through Kossel's octet theory, according to which the atoms in the bonding state (of the crystal lattice) strive for a "noble gas configuration" in their outer electron shell. - Although W. Kossel was aware of the work of Erwin Schrödinger and others on the quantum theory of atomic structure and chemical bonding, he cemented "his" valence theory of chemical bonding on the basis of given bond polarities and with the exclusion of the new quantum theoretical views of covalent structures in molecules: From a non-quantised axis of binding energy only ions can be formed.

8.6 Consequences of the ion and valence theory

Since S. Arrhenius and supported by W. Kossel, organic chemical thinking has also been permeated by polar, ionised or temporary ionisable structures. Terms such as "nucleophilic" or "electrophilic" often bear witness to this without having defined the polarizability and boundary structures of the reactants in the case of substitutions. However, terms such as "hydride" ions, "hydride" migration, "hydride" shift, "carbo"-cations, "carb"-anions are particularly problematic, when these are essentially transferred from purely preparative chemistry to structures of a pH-specific physiological chemistry and appear in publications. Homosynthesis and/or homolysis of a chemical bond presuppose the existence of free radicals as electroneutral, i.e. uncharged (possibly polarised) molecules, even if these originate from the cleavage of polar bonds. The existence of radicals is based on quantum chemistry. Radicals are fundamental both to understanding becoming and decay and to seeing changes in the environment and atmospheric chemistry. With all due respect to scientific achievement, this sometimes casts shadows on developments "afterwards": **Arrhenius-Kossel's dilemma** leaves no room for free radicals. To this day, it manifests itself in an uncritical weighting of molecular bond polarities and electrostatics in research and teaching - also in the affected sciences. Thus, the "citrate cycle" is formulated via anions of hardly dissociating organic acids ⁸⁹.

8.7 Air pollution, health and climate ⁹⁰

Primary air pollutants are particles of different size, shape, density, composition and reactivity in different phases that are introduced unchanged into the air and remain there until they sediment. Examples are dust, smoke and soot particles, i.e. solids, liquid substances, exhaust gases, odorous substances and aerosols.

Among them are main components of air pollution (mostly of anthropogenic origin), which prove to be pathogenic primarily by their size and thus according to their depth of penetration into the lungs.

1. When approx. 50% of the fine dust particles have a diameter of 10 μm , they are categorised as "coarse fine dust" (PM_{10}). PM means particulate matter ⁹¹. Such particles have almost cell size, penetrate deep into the bronchial tubes and initiate inflammation by activating the phagocytic defence.

2. Smaller particles below 2.5 μm ($\text{PM}_{2.5}$) reach the size of bacteria. They get into the alveoli, where at the end of the respiratory tract an exchange of O_2 for CO_2 takes place between alveolar spaces and the blood cells of the capillaries. Pollutants of the size mentioned are considered "fine particulate matter".

3. Below PM_1 are air pollutants with ultrafine particles ($\text{PM}_{0.1}$ of 100nm diameter), i.e. nanoparticles. - Ultra-fine pollutants and nanoparticles ⁹² pass the blood-air barrier. They enter all organs, cells and cell nuclei via the circulatory system. - Secondary air pollutants are formed by chemical changes ⁹³ of particles in the atmosphere, such as from ammonia by hydrochloric acid gas ($\rightarrow\text{NH}_4\text{Cl}$). Vehicle functions of air pollutants are of importance, both with their surfaces for chemical reactions and through the transport of toxins or allergens. Direct air pollutants are also flower dusts. They are triggers of pollen allergies. - A new Swiss review article ⁹⁴ throws vivid light on the interrelationships and practical effects of air pollution, climate and allergies. With the absence of cold winters for many years, the greenhouse effect favours an earlier and longer-lasting release into the atmosphere of ever

greater quantities of highly allergenic pollens such as birch, ash, hazel and grass pollen. The large-scale SAPALDIA ⁹⁵ study (published in 1995), which has been in progress since 1991, shows an increase in the severity of pollinosis and lung diseases in adults in middle to old age, as well as an increase in food allergies due to cross-reactions between some pollen allergens and foods (e.g. nuts, fruit and some vegetables).

The SCARPOL study ⁹⁶ published in 1997 documents a significant increase in juvenile allergic, predominantly respiratory symptoms between 1986 and 1994: - Allergies continue to increase according to air pollution with traffic and towards large cities, where the limit values of nitrogen oxide ($90\mu\text{g}/\text{m}^3$) and ozone ($120\mu\text{g}/\text{m}^3$) are often exceeded. Children in particular react sensitively to ozone pollution with respiratory diseases (asthma and rhinitis) - occurring more frequently in the vicinity of traffic junctions ^{94,97}.

Increasing traffic and air pollution also induce "plant stress", which manifests itself in part through increased synthesis of pollen allergens and pollen-associated lipid mediators (PALM) with pro-inflammatory and leukotriene-like properties ⁹⁸. - Therapies for allergy-related diseases are becoming successively more difficult. Furthermore, not only PM₁₀ particles but also finer particulate matter as well as nanoparticles and pollen themselves prove to be carriers of particulate matter from tyre wear, soot, metal oxides, O₃, nitrogen oxides, CO, toxins and carcinogenic adhesive substances ⁹⁸.

8.8 Climate-relevant coolants and propellants ⁹⁹

The Federal Environment Agency (UBA) published August 2019 in < CLIMATE CHANGE 08/2010 >: Avoiding fluorinated greenhouse gases - ways to phase out ¹⁰⁰: To protect the ozone layer, chlorofluorocarbons (CFCs) and halons (halogenated hydrocarbons in prohibited in fire extinguishers). These highly effective refrigerants were quickly replaced by the use of semi-hydrofluorocarbons (HFCs), which allegedly do not damage the ozone layer. They are now used in freezers, air conditioning, refrigeration, firefighting, insulation foaming and often as medical spray propellants. Together with per-fluorinated hydrocarbons (PFCs), sulphur hexafluoride (SF₆) and nitrogen trifluoride (NF₃), these refrigerants are declared as F-gases (in special applications). They have also proven to be highly effective greenhouse gases that further heat up the climate ⁹⁹. Their contribution to the total emission of climate-damaging gases is currently 2% worldwide and is estimated to be around 6% by 2050 - with an eye to the future - due to the "increasing demand" in developing countries. According to the UBA article (see foreword), F-gases are 100 - 24000 times more harmful to the climate than carbon dioxide. - It has been estimated that in 2050 about 80% of F-gas emissions will come from stationary areas (i.e. industry, rooms, supermarkets) and mobile refrigeration and air-conditioning applications: They will thus also be released from car-, bus-, train- and ship air-conditioning systems. The above-mentioned 80% then allegedly corresponds to 3211 mega-tonnes of CO₂ equivalents.

9. Water treatment and disinfection

9.1 Water treatment and improvement of water quality

A rapidly increasing world population is currently leading to alarming geopolitical migration problems. At the same time, falling water tables due to climate change are making the availability of clean water a matter of survival, not only in underdeveloped countries. Contaminated drinking water was already proven in 1883 by Robert Koch to be the cause of cholera epidemics ¹⁰¹. The future clearly lies in the reuse of water. According to *planet wissen*, the Singapore water utility is a global pioneer in the production of highly purified drinking water - called "New Water" - from municipal wastewater. This is achieved through a multiple system of high-performance membranes through which the wastewater is pressed. In the process, bacteria are filtered out, among other things. In addition, the "recycle wastewater" provides for a parallel activated intensive UV light disinfection, which reduces the remaining germs in the water to a maximum ¹⁰². An unsolved problem is the removal of drug residues and hormone preparations, which enter the wastewater in very low but persistent concentrations. The industry is obliged to develop detectable and easily degradable alternatives to the existing products.

9.2 Calcium hypochlorite - chlorine - chlorine dioxide - ozone ¹⁰³⁻¹⁰⁵

In July 2018, the Federal Environment Agency published a list of treatment substances and disinfection processes in accordance with § 11 of the Drinking Water Ordinance ¹⁰³. For oxidative disinfection of most water germs, sodium hypochlorite pentahydrate in chlorine bleaching lye ($\text{NaOCl}\cdot 5\text{H}_2\text{O}$), calcium hypochlorite $\text{Ca}(\text{OCl})_2$ among others in the technical chlorinated lime of agriculture, chlorine gas (Cl_2) for bathing water, chlorine dioxide (ClO_2) mainly for drinking water treatment and ozone (O_3) (produced in special "ozonisers") for water and drinking water treatment are used. Recommended additions of free chlorine or ClO_2 and O_3 are given. Reference is made to reaction products that must be taken into account. The disinfecting substances are also listed in the publication of the <Zweckverband der Wasserversorgung Nordostwürttemberg (NOW): "Trinkwasseranalysen - Mittelwerte 2019/20">. The table of the treatment substances used to disinfect the water additionally notes the UV irradiation ¹⁰⁴. Chemical disinfection by the treatment substances mentioned proceeds via combinations of radical and ion mechanisms in the aqueous milieu that are difficult to trace. - Depending on the pH value, transparency of the abiotic environment, molecularly variable polarizability of the particles, pressure and temperature, the biologically hardly specifiable germs are probably mainly exposed to radical reactions. Presumable processes during chlorine gas disinfection are:

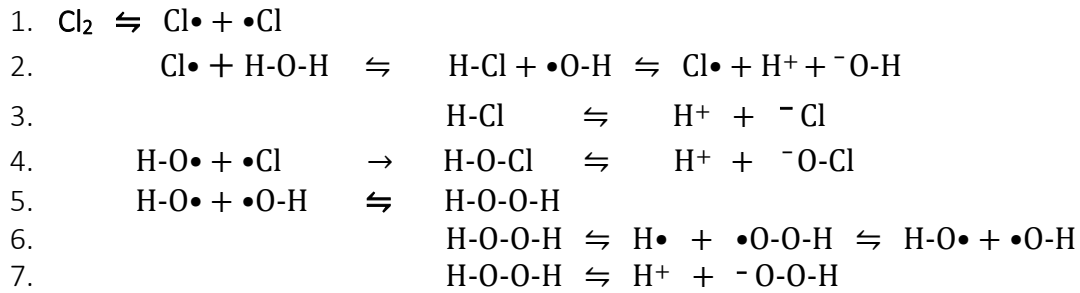


Fig. (15) Possible radicals and ions as abiotic reactants of disinfection*

*Note: Direct radical effects such as chlorination, peroxidation and/or ions in saponification probably contribute to the destruction of germs in their DNA. Radical formation may be promoted by the formation of molecular hydrogen bonds.

9.3 Treatment of water from emergency wells - drinking water disinfection tablets ¹⁰⁵

For the needs of the German Armed Forces, in case of defence and for disasters in civilian areas, 2008 treatment and disinfection processes for water with tablets of sodium-dichloro-isocyanurate (1,3-dichloro-6-hydroxy-1,3,5-triazine-2,4-dione as dihydrate) enlarged (water tablets).

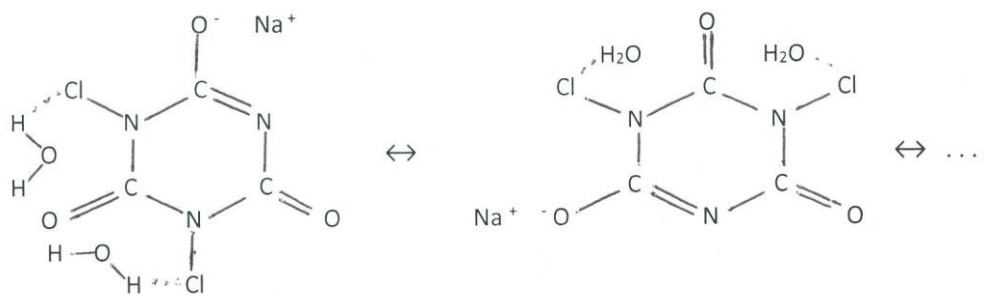


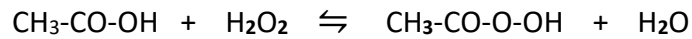
Fig. (16) Sodium dichloro-isocyanurate*

*Note: The nitrogen-chlorine bond is destabilised in the dihydrate by H-bonds. This is where the smell of chlorine comes from.

9.4 Peroxyacetic acid (peracetic acid): Properties and applications ¹⁰⁶

Peroxyacetic acid (PES) is one of the most important agents for water and wastewater disinfection with high material compatibility, also in disaster management. It simplifies and supplements sterilisation processes. In the medical field, for example, it has been shown to be effective in skin and hand disinfection, surface disinfection, room and *ambient air disinfection*, the disinfection of surgical instruments and protective clothing, and its use as an antiseptic, all with low, controllable toxicity. Peroxyacetic acid is used in veterinary medicine for epidemics and for the germ-free rearing of animals. - The use of alkalisated PES is becoming increasingly important in the food industry to reduce the infestation of e.g. tomatoes and fruit with harmful yeasts and moulds. - All applications take place at relatively low PES concentrations in aqueous solutions or aerosols and can be realised at low temperatures if necessary. Resistance to PSE is unknown. PES does not induce allergies and is not carcinogenic. Disinfection is surprisingly fast. It is environmentally friendly. Its effectiveness depends on the structure of the surfaces, concentration and exposure time.

After extensive research, there is no real alternative to disinfection with peracetic acid. It is listed by the RKI in accordance with §18 of the Infection Protection Act and is therefore also approved in accordance with the German Medicines Act. PES is formed in an equilibrium reaction of acetic acid with hydrogen peroxide:



The pure preparation of the original substance in 1912 proved to be problematic because of the danger of explosion at higher temperatures. Diluted PES solutions were then said to be dangerously unstable without justification. - Peter Bützer published a comprehensive *review article on peroxyacetic acid in 2012*: As a simple but very effective agent, it is the most important organic peroxide and micro-biocide for the disinfection of all known bacteria, viruses, yeast cells, fungi and spores¹⁰⁷. Research into PES began (without knowledge of the reaction products) as early as 1821 in France. Since about 1950, interest has been directed towards the problems of decontamination in military medicine and practical disaster control. The biological effects of PES differ from those of the components CH₃COOH and H₂O₂. Bützer is one of the few authors to see the oxidative effects of peroxyacetic acid primarily in the formation of reactive radicals and not in the release of atomic oxygen or the splitting off of (singlet oxygen).

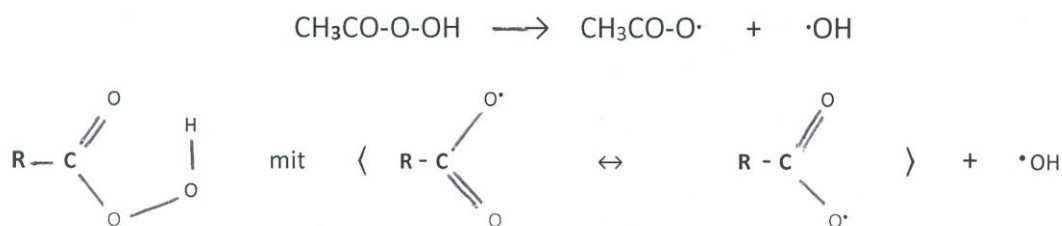


Fig. 17 Functional group of peroxyacetic acid (with resonance-stabilised acetoxy radical) *.

*Note: The structure of the functional group was taken from IR spectroscopic studies in 1952. It makes the formation of reactive radicals understandable via resonance¹⁰⁸.

10. *Pneumonia prophylaxis*¹ - literature sources^{109,110} - radical chemical aspects

Pneumonia is a bacterial, acute or chronic inflammation of the lung tissue. It is a frequent cause of death among infectious diseases. In *atypical pneumonia*, viruses are the most common pathogens. *Pneumonia prophylaxis* includes preventive measures for patients to avoid this pneumonia. These include improving breathing by removing bronchial secretions, humidifying as well as disinfecting the room air and optimising the breathing air. Obstructive pulmonary diseases (COPD) also develop without bacteria.

10.1 Inhalation prophylaxis by disinfection:

In a brochure entitled "Room air disinfection with peracetic acid (PES) aerosols in intensive care units and its consequences for the treatment of the hospital respiratory problem", Kesla AG gives examples of the use of Wofasteril in recumbent patients with severe respiratory diseases¹¹¹. The preparation convinced the medical officers of the German armed forces: it

was included in the disaster medicine disinfectant list ^{106,107}. - In the discussion of the aforementioned article ¹¹¹ an improvement in the clinical picture of some patients who asked for further treatment with nebulised peracetic acid was reported. - At correctly calculated concentrations, no de facto adverse health effects occurred. A 1% PES solution with an ethanol content of 30% up to a concentration of 4.6 mg PES/m³ was distributed homogeneously in the room air as an aerosol by ultrasonic fogging (standard). With this procedure, the airborne germ content in patient rooms was demonstrably reduced practically completely. Thus, all known germs were largely destroyed ¹¹¹.

In addition to the entry of peroxyacetic acid in the *disinfectant list of the German Federal Armed Forces* and the already mentioned review paper by P. Bützer: < *Peracetic acid, simple but effective* >, radical chemical aspects are convincing in inhalative pneumonia prophylaxis: 1. by *ultrafast dimerization of the hydroxyl radicals of viral inflammation genesis* with •OH-radicals from peroxyacetic acid to H₂O₂ as well as 2. by the longer-acting "*broadband destruction*" of viral DNA or viral RNA with capsids* ¹¹² or larger, enclosing viral envelope proteins by the indicated resonance-stabilised alkoxy radicals ^{107,108}.

* Note: Capsids are rod-shaped or spherical protein shells of the nucleic acids of infectious viruses.

10.2 Dexamethasone -type therapy:

Respiratory patients suffering from coronaviruses (SARS-COV-2 or Covid-19) were able to survive significantly longer with concomitant dexamethasone therapy. The partial and spectacularly cost-effective success in this life-threatening disease was recently confirmed by a study of the "RECOVERY Collaborative Group" P. Horby et al. with <*Dexamethasone in Hospitalized Patients with Covid-19 - Preliminary Reports*> ¹¹³. It led to *recommendations for intensive care therapy of patients with Covid-19* according to the S2k guideline and the Robert Koch Institute ^{114,115} including dexamethasone.

Budesonide as an inhaled corticosteroid is likely to have a comparable effect: In severe asthma and chronic obstructive pulmonary disease (COPD), a β_2 -sympathomimetic (also inhaled) is often administered beforehand to dilate the bronchial tubes, whereby terbutaline sulphate* has proven successful.

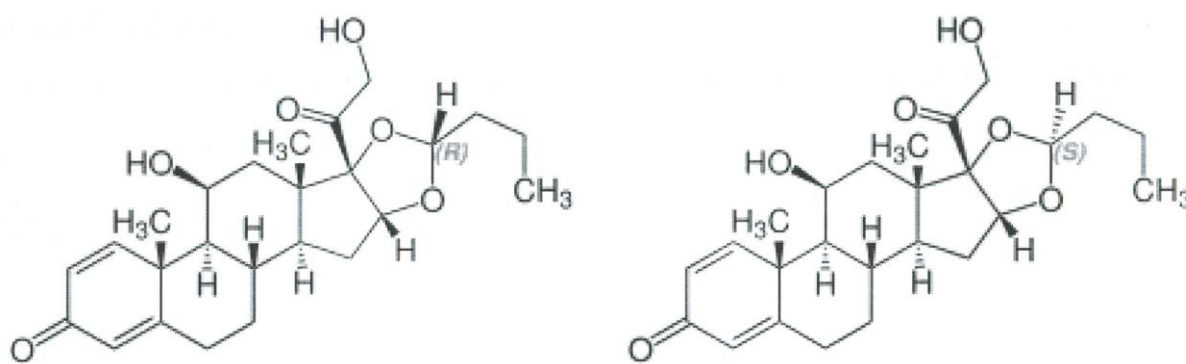


Fig. ⑱ Budesonide (e.g. Pulmicort®) a halogen free inhaled synthetic glucocorticoid.

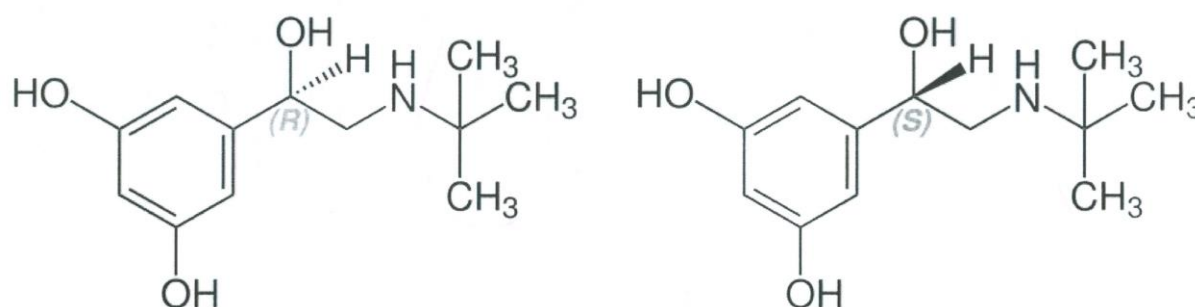


Fig. ⑲ Terbutaline sulphate (e.g. Aerodur®) an inhalable β_2 -sympathomimetic

* Remark:

The protons in terbutaline sulphate are localised at the nitrogen. - Figures 18 and 19 were taken from WIKIPEDIA 2021

Anti-inflammatory effects of dexamethasone as a "direct" radical scavenger have been theoretically proven by published mechanisms for the disposal of HO•- radicals to hydrogen peroxide³⁴. Compare, however, recent attempts to explain the anti-inflammatory effects of glucocorticoids purely enzymatically¹¹⁶. However, no metabolite comparable to the acetoxy radical^{107,108} of peroxyacetic acid is formed in the exemplary scavenger reactions^{30,34} that could kill coronaviruses themselves. This may explain the limited effect of dexamethasone therapy and comparable strategies in more severe covid-19 illnesses.

10.3 Selenium in viral infections and covid-19

In a controlled study in 1998-2000, 186 HIV-positive people were given 200 µg/d selenium (as selenium yeast monthly adjuvant controlled for 2 years)¹¹⁷. - Result: Hospitalisation times as well as therapy costs decreased in the selenium group also due to the simultaneous decrease in the frequency of concomitant and spontaneous infections. However, CD4 was not significantly changed in either arm.

Selenium, Selenoproteins and Viral Infection (L. Chavatte et al.) in Nutrients 2019^{118a}: This is a large-scale review with 241 cited partly recent publications! When viral infections are associated with the generation of highly excess oxygen radicals (ROS), oxidative stress may become the main feature of the disease for patients. Oxidative stress itself promotes viral replication, which in turn enhances the overall pathogenesis. Clinical symptoms of virally ill patients, who usually have a nutritional state with detectable selenium deficiency, are often accompanied by severe inflammation. - In biology and medicine, selenium is considered an essential trace element with antioxidant and anti-inflammatory properties of some of its compounds. Selenium is incorporated into proteins in the form of selenocysteine. Very important antioxidant enzymes are glutathione peroxidases and thioredoxin reductases. These selenium-proteins are supporting pillars of redox biology, in which the physiological flow equilibrium between a necessary and excessive production of reactive oxygen and nitrogen species is to be maintained. - The importance of selenium deficiency began with the discovery of Cocksackie virus as a characteristic feature of Keshan disease, an endemic cardiomyopathy in north-eastern China.

According to the above work^{118a} the viruses allegedly "induce" enzymes that "generate" reactive oxygen species and "interfere" with antioxidant defence mechanisms. For example, *xanthine oxidase*, which contributes to the degradation of purine bases, "produces" hydrogen peroxide (H₂O₂). Viral pathogens are involved in the production of reactive oxygen species in other infections: for example, the human immunodeficiency virus (HIV), the hepatitis B virus (HBV), the hepatitis C virus (HCV), the Epstein-Barr virus (EBV), the herpes simplex virus1 (HSV-1), the respiratory syncytial virus (RSV), the human T cell leukaemia virus type 1 (HTLV-1) and influenza viruses. Some viruses are known from diseases experienced or from information provided by doctors. All the viral diseases listed are accompanied by more or less severe inflammation. At the end of the publication, the authors discuss seleno-proteins in viral genomes. It seems likely that such seleno-proteins, e.g. in the molluscipoxviruses (trigger of smallpox skin diseases) or in fowl-pox viruses, protect the viral genomes themselves from radical destruction and thus ensure their resistance to their own degradation during prolonged survival. Presumably, such protective mechanisms through seleno-proteins also exist in other virus genomes. The conclusion of the work is: „The

mechanisms of ROS generation by various viruses are diverse, but in several cases the host antioxidant defence enzymes and especially members of seleno-proteome, are targeted". Selenium status marks the severity of COVID-19 ^{118b}.

The journal *Nutrients 2020* also published: *Selenium Deficiency is Associated with Mortality Risk from COVID-19 (Schomburg et al.)* ¹¹⁹. The paper also discusses the often fatale pneumonia outbreak towards the end of 2019, which rapidly developed into a pandemic starting in the city of Wuhan (Hubei region, China) and triggered by highly infectious coronaviruses (SARS-CoV-2 viruses). - The authors state: "The mortality risk from a severe disease like sepsis or polytrauma is inversely related to Se status. We hypothesised that this relation also applies to COVID-19." – The assumption is justified by analyses of referential values of prospective studies by the authors on selenoprotein P (a selenium carrier protein) in patients with colorectal cancer in comparison with volunteers (so-called "EPIC study"): 166 serum samples were collected in sequence from 33 COVID-19 patients. *First biomarker*: Serum selenium content was analysed by total reflectance X-ray fluorescence spectroscopy (TXRF). *Second biomarker* was selenoprotein P: Selenoprotein P (SELENOP) is under-expressed in poor selenium status. The analysis of selenoprotein P in COVID-19 patients was performed according to the cited EPIC analysis using a validated and commercial immunological test method for the specific detection of selenoprotein P (sandwich method: measurement of monoclonal antibodies against human selenoprotein P). *Third marker*: The activity of glutathione peroxidase-3 was determined. - The COVID-19 patients were differentiated according to gender, age and concomitant previous diseases on the one hand and death and discharge on the other. The study methods, which were generally based on our own preliminary work, were subjected to statistical analyses. Closer linear correlations were found between 1. the sample concentration of selenoprotein P and serum selenium, 2. glutathione peroxidase and serum selenium, and 3. glutathione peroxidase and the selenoprotein P content of the samples. - For the COVID-19 patients showed significantly worse selenium status, selenoprotein P expression and glutathione peroxidase in relation to the EPIC analysis. The Markers of the patients who eventually died were significantly worse than those of survivors in whom selenium levels subsequently normalised. For the authors the examination of selenium status provides important diagnostic information. It promotes the discussion about additional selenium administration in critically ill patients and patients with selenium deficiency. However, the cause of the diagnostic significance of selenium for the recovery of COVID patients remains unknown "against the background of the character of a pure observation" in this study. –

The above work is supported by a more recent review article from China (study leader: M. P. Rayman) in the journal *Redox Biology* ¹²⁰. - Are antioxidant seleno-proteins themselves protective components of the genome of covid-19 viruses? - This question is of great importance. This study by J. Zhang, E.W. Taylor, K. Bennett, R. Saad and M. Rayman should also be mentioned for another reason ¹²⁰. In it, the course of Covid-19 disease is linked to the regional selenium content (cf. 118b): the severity of the disease and mortality is pronounced in selenium-poor regions. In the Hubei region around Wuhan, which is very rich in selenium, a milder course of the disease and faster recoveries have been observed.

10.4 Problems with the approval of medicines

Recent publications on COVID-19 often have the advantage of justifying their statements experimentally and systematically. In contrast, however, < Selenium, Selenoproteins and viral infection > ^{118 a} gives examples, circumstantial evidence and sobering statements on possible causes of disease.

Attempts to market selenium preparations as medicines have been known for years.

Regulatory authorities for medicinal products are generally not responsible for nutritional issues and food supplements. However, the latter may be used as "adjuvants" in therapy studies if necessary. They then do not claim a cure themselves but a reproducible alleviation of symptoms. - Fortunately, there are control bodies to check whether a propagated substance alleviates characteristic symptoms of diseases for defined indications according to the German Medicines Act, or whether it significantly prolongs the lives of seriously ill patients, or whether it even enables healing. Suppliers of selenium-containing substances also have to face such questions.

As a rule, non-medical reasons should have no influence on the approval of a substance as a drug. Suppliers of selenium pharmaceuticals are therefore well advised in their own interest to first clarify and analyse poly-pragmatic strategies (e.g. the number of components during multiple diseases with possible interactions in an infusion) with the investigators involved and to minimise them as far as possible in consultation with them in order to then be able to seriously document the efficacy of *the "mono-substance" to be tested*. This also includes the adjuvant administration of seleno-methionine or selenium yeasts separated from the main therapy. - Chronobiological problems should also be discussed. In the case of a marketing authorisation application, extended justifications should prove helpful, especially for authorities. However, our marketing authorisation bodies for medicinal products must then ask themselves with regard to the selenium problem: Is selenous acid (or sodium selenite solution) not a medicinal product only because selenium is an essential trace element? - Or does a persistent refusal to accept radicals as a source of inflammation reveal a basic chemical training of our medical decision-makers that needs to be very critically reconsidered and improved?

11. Only infection prophylaxis or also drug therapy?

The objective of any infection prophylaxis* for healthy people or patients of any age is to prevent infections by pathogenic viruses, bacteria and fungi. What have prophylactic vaccinations achieved against viral infections so far?

As early as 1988, Sammalkorpi et al. published the drop in serum selenium levels in various acute viral and bacterial infections ¹²¹. However, a bundle of medicinal measures should also be used to therapeutically successively destroy the detected germs and thus prevent contagious transmission to other people. - Antibiotics are the treatment of choice, e.g. against pneumococci (*Streptococcus pneumoniae*). Without them, bacterial pneumonia, infectious asthma and chronic obstructive pulmonary disease (COPD) would often worsen dramatically. Non-infected subjects with a supposedly balanced diet but a nevertheless proven selenium deficiency could benefit from appropriate dietary supplements. But why the Covid-19 virus was able to spread so quickly in the selenium-rich Hubei region (starting from the metropolis of Wuhan) would have to be determined via the genome of this dangerous and apparently long-lived germ. It would be necessary and of general interest to find out, irrespective of the environmental pollution of this large industrial city.

*Note: An inhalation application of selenous acid ($\text{Na}_2\text{SeO}_3 \cdot 5\text{H}_2\text{O}$) by ultrasonic nebulisation** could ensure a fast-acting, anti-inflammatory measure against already existing germs, both prophylactically and therapeutically. The same applies to infusions.

** Ultrasound nebulisers are manufactured, for example, by Pari (Munich)

On the one hand, in the case of an unsatisfactory selenium status, selenium blood values and Se tissue levels are also normalised via dietary selenium and a possible viral load is at least partially lowered. On the other hand, primarily only a systemic application of selenic acid reduces radicals of viral or bacterial origin by direct disposal comparable to dexamethasone, even in cases of severe inflammation (in contrast to selenium yeasts and organic selenium compounds) ^{30,34}.

If antioxidant self-protection of unknown viral genomes against free radicals from the host organism cannot be ruled out in principle, further therapeutic approaches (if indicated also inhaled) should first be considered. Already addressed were:

Budesonide - peroxyacetic acid (peroxy-citric acid or other peroxy-carboxylic acids) - selenous acid as ($\text{H}_2\text{SeO}_3 \cdot 5\text{H}_2\text{O}$) – dimethylsulfoxide - derivatives of chloric acid, permanganic acid and ozone. The fluorine-containing dexamethasone is not suitable for long-term therapy of chronic respiratory diseases.

For the time being, serious results on the specific medical effects of the extremely costly and politically promoted vaccination campaigns in the Corona pandemic remain to be seen.

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Anmerkungen des Autors zu

“Fragmente der Katastrophenmedizin IV
Radikale in unserer Natur und die Natur der Radikale“

Die veröffentlichte deutsche Fassung enthält umständehalber einige Fehler durch ausgelassene Satzteile sowie grammatikalische und orthographische Fehler. Diese wurden vom Autor in der englischen Version weitgehend verbessert.

Dabei ist auch darauf hinzuweisen, dass die Umwandlung neuer oder nicht gängiger interdisziplinäre Begriffe aus der Forschung von amerikanischem Englisch in britisches Englisch eine gewisse Zeit erfordert.