

SUPPLEMENTARY INFORMATION

Zinc(II) - the overlooked éminence grise of chloroquine's fight against COVID-19?

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Table S1. Summary of non-CQ and non-HCQ based therapeutics in anti-COVID-19 clinical trials.

Drug	Therapeutic effect	Reference
<i>Acetylcysteine</i>	mucolytic drug used in acute and chronic bronchitis and other respiratory diseases with high mucosal secretion; moderates clinging mucous secretions and enhanced glutathione S-transferase activity	[1]
<i>Amlodipine</i>	calcium channel blocker; indicated for the treatment of high blood pressure and chronic angina pectoris and angiospastic angina	[2,3]
<i>Amoxicillin</i>	β -lactam antibiotic; used to treat a number of bacterial infections such as acute sinusitis, acute cystitis and acute pyelonephritis	[4]
<i>Anakinra</i>	recombinant interleukin-1 receptor antagonist, immunosuppressor; used in rheumatoid arthritis together with methotrexate when self-methotrexate treatment response is insufficient; studies have shown that administration of anakinra reduces 28-day mortality by 30% in patients with severe sepsis and macrophage activation syndrome	[5]
<i>Acetylsalicylic acid (Aspirin)</i>	nonsteroidal anti-inflammatory drug with analgesic, antipyretic and anti-inflammatory effect, also platelets aggregation inhibitor; exhibits antiviral activity against influenza virus (NCT04343001); 9 CTs (in mono- or combination therapy with, e.g. clopidogrel, rivaxoban, ivermectin, losartan)	[6,7]
<i>Atorvastatin</i>	HMG-CoA reductase inhibitor, hypolipemic drug; used for lowering cholesterol and in prevention of cardiovascular disease	[8]
<i>Atovaquone</i>	synthetic hydroxynaphthoquinone with antiprotozoal activity; used for the treatment of malaria in combination therapy with proguanil hydrochloride	[9]
<i>Aviptadil</i>	synthetic form of vasoactive intestinal polypeptide (VIP); used for the treatment of ARDS; very promising in the treatment of COVID-19 patients with associated acute respiratory distress (NCT04311697)	[10]
<i>Azithromycin</i>	macrolide antibiotic; induces antiviral response in bronchial epithelial cells and may also be used to treat malaria; may be helpful in treating coronavirus infections	[11,12]
<i>Baricitinib</i>	Janus kinases inhibitor; used for the treatment of rheumatoid arthritis as monotherapy or associated with methotrexate; in severe acute respiratory syndrome coronavirus 2 infection with ongoing pneumonia may decrease the hyperinflammation (prevention of lung damage)	[13–16]
<i>BCG Vaccine</i>	vaccine against tuberculosis; <i>in vitro</i> and <i>in vivo</i> studies have shown a beneficial effect also against various viral pathogens, e.g. respiratory syncytial, yellow fever, herpes simplex and human papilloma viruses	[17]
<i>BLD-2660</i>	inhibitor of calpain (CAPN) 1, 2, and 9; has a potential in reducing viral replication and nonproductive host-response to infection (the cause of morbidity and mortality in COVID-19), NCT04334460	[18]
<i>Bromhexine</i>	mucolytic drug; used for the treatment of respiratory disorders; has also a potential for the treatment of influenza virus and coronavirus infections. In a	[19]

	clinical trial (NCT04340349), it will be tested together with hydroxychloroquine (in low doses)	
<i>Budesonide</i>	glucocorticosteroid; used in bronchial asthma treatment, acute laryngitis, tracheitis, bronchi, also in chronic pulmonary disease to reduce inflammation; can suppress the immune reaction of the respiratory system locally	[20,21]
<i>Calciferol</i>	vitamin D; it is proposed that vitamin D, as a nutritional ergogenic aid, could be a potential intervention to fight against COVID-19 infected patients; bovine coronavirus studies have shown that reduced levels of vitamin D in calves have contributed to their infection (NCT04334005)	[22,23]
<i>CamostatMesilate</i>	protease inhibitor; used in remission of acute symptoms of chronic pancreatitis and postoperative reflux esophagitis; acts as an inhibitor of the TMPRSS2 host cell serine protease, which is needed to prepare viral S protein for entry into the cell (NCT04338906); reduces significantly the infection of Calu-3 lung cells by <i>SARS-CoV-2</i> (in vitro study)	[24,25]
<i>Captopril</i>	zinc ion-utilizing human angiotensin converting enzyme (ACE); mainly used to treat hypertension and kidney problems caused by diabetes	[26]
<i>Ceftarolinefosamil</i>	broad-spectrum oxyimino cephalosporin antibiotic with activity against respiratory pathogens including <i>S. pneumoniae</i> , <i>H. influenza</i> , <i>M. pneumoniae</i> , and influenza/parainfluenza viruses; used in non-hospital pneumonia	[27,28]
<i>Ceftriaxone</i>	widespectrum antimicrobial activity against both gram-positive and gram-negative organisms	[29]
<i>Chlorhexidine</i>	disinfectant and antiseptic agent; used e.g. as skin disinfectant; strong bactericidal and bacteriostatic effect; some <i>Pseudomonas</i> , <i>Proteus</i> , acid-resistant mycobacteria and spores are resistant to this agent	[30]
<i>Chlorine dioxide</i>	disinfectant; strong inhibitory effects on microbes and parasites	[31]
<i>Ciclesonide</i>	inhaled corticosteroid; used in chronic bronchial asthma; shows good antiviral activity against <i>SARS-CoV-2</i> ; used also in mild <i>SARS-COV-2</i> infections in combination with hydroxychloroquine (NCT04330586)	[32]
<i>Clevudine</i>	antiviral drug; activity against hepatitis B virus (HBV)	[33]
<i>Cobicistat</i>	CYP3A inhibitor with no antiviral activity; indicated for <i>HIV</i> ; used as a pharmacokinetic enhancer of certain antiviral agents (e.g. elvitegravir, atazanavir, and darunavir)	[34]
<i>Colchicine</i>	plant alkaloid; has some effect on the inflammasome NLP3 and microtubule formation which are associated with the pathogenetic cycle of coronavirus (NCT04326790)	[35,36]
<i>Cyclosporine</i>	immunosuppressant; used to help prevent rejection in organ transplant patients; it is also used to treat rheumatoid arthritis and psoriasis. In a clinical trial, it will be added to standard treatment in order to improve the prognosis of hospitalized patients with <i>SARS-COV-2</i> infections. The hypothesis of clinical study is that the cyclosporine is safe and it will have antiviral and anti-cytokine effects (NCT04412785)	[37,38]
<i>Darunavir</i>	protease inhibitor; used to treat HIV associated with ritonavir and cobicistat	[39]
<i>DAS181</i>	recombinant protein; antiviral agent; <i>in vitro</i> and <i>in vivo</i> studies have shown against parainfluenza and other viruses activity	[40–42]
<i>Deferoxamine</i>	chelator of ferric ion; used to treat transfusion related chronic iron overload; inhibits human <i>cytomegalovirus</i> replication	[43,44]
<i>Defibrotide</i>	oligonucleotide mixture with demonstrated antithrombotic, fibrinolytic, anti-adhesive and anti-inflammatory actions; used for hepatic veno-occlusive disease (VOD)	[45]
<i>Dexamethasone</i>	corticosteroid; anti-inflammatory, anti-allergic and immunosuppressive properties; also in chronic corticotherapy; it is being investigated for the treatment of COVID-19 due to the safe and effective activity of corticosteroids (also dexamethason) in the prevention of acute respiratory distress syndrome (ARDS), including viral pneumonia (NCT04327401)	[46,47]
<i>Diltiazem</i>	calcium channel blocker; used to treat hypertension, angina pectoris and arterial	[48–50]

	hypertension; according to the data from Italy/USA, acute myocardial infraction with imposed respiratory failure significantly complicates the course of disease; used in the combination with thiazide, ACE inhibitors and ARB (NCT04330300)	
<i>Doxycycline</i>	strong and broad-spectrum inhibitor of matrix metalloproteases (MMPs); tetracycline-class antibiotic; used to treat bacterial infections, e.g. acne or urinary tract infections; may have also a role as an effective anti-viral agent and as an anti-inflammatory drug. Because it is know, that (i) the coronaviruses bind to metalloproteases (MMPs) of the host and (ii) the tetracyclines chelate the zinc from MMPs and their chelating activity can help inhibit COVID-19 infection by limiting of ability to the <i>SARS-CoV-2</i> virus replication, the primary aim of this study is to test whether doxycycline can benefit patients with severe acute respiratory syndrome coronavirus 2 infection (NCT04433078)	[51–53]
<i>Eculizumab</i>	monoclonal antibody, terminal complement inhibitor; used for treatment of atypical haemolyticuraemic syndrome and paroxysmal nocturnal haemoglobinuria	[54]
<i>Enoxaparin</i>	low molecular weight heparin; anticoagulant; used in angina pectoris and cardiac infraction; may play an anti-inflammatory role by reducing the release and biological activity of IL-6; investigated also as a preventing agent against <i>SARS-CoV-2</i> infection (NCT04345848)	[55–59]
<i>Escin</i>	mixture of saponins; has anti-inflammatory and anti-edematous effects; usually used to treat e.g. hemorrhoids, varicose veins, hematoma or venous congestion	[60]
<i>Favipiravir</i>	antiviral agent; selective and potent inhibitor of influenza viral RNA polymerase; used for the treatment of influenza; has a high potential activity against <i>SARS-CoV-2</i> virus; a few combination therapy CTs are actually performed with e.g. tocilizumab (NCT04310228), imatinib (NCT04356495), nitazoxanide and chloroquine (NCT04345419), hydroxychloroquine, azithromycin and zinc sulfate (NCT04373733)	[61,62]
<i>Fluvoxamine</i>	anti-depressant; used to treat obsessive-compulsive disorder; proposed to be used early in the course of the <i>SARS-CoV-2</i> infection to prevent more serious complications like shortness of breath (NCT04342663)	[63]
<i>Folic Acid</i>	type of vitamin B; used for production and maintenance new cells in the body and for prevention of the changes in DNA that can lead to cancer. It is suggested that folic acid may be used to inhibit the furin enzyme and may be useful in preventing or treating COVID-19 associated respiratory disease in the early stages of the disease	[64]
<i>Galidesivir</i>	antiviral drug; <i>in vitro</i> studies have shown activity against many viral pathogens and infectious agents including <i>MERS-CoV</i> and <i>SARS-CoV</i>	[65]
<i>Giapreza</i>	angiotensin II; used for increase blood pressure in adults with septic or other distributive shock	[66,67]
<i>Hydrocortisone</i>	corticosteroid; used for the treatment of e.g. allergic disorders, severe asthma skin conditions, ulcerative colitis, arthritis, lupus, multiple sclerosis, autoimmune etiology diseases, or lung disorders; in used in combination therapy with some antimicrobial agents, interferon beta and immunosuppressors (NCT02735707)	[68,69]
<i>Ibuprofen</i>	non-steroidal anti-inflammatory drug; used for relief of rheumatic or muscular pain, back pain, and the pain of non-serious arthritic conditions, caused by swelling, stiffness and inflammation; evaluation of the reduction in severity and progression of lung injury in patients with <i>SARS-CoV-2</i> infections is under investigation/4 th phase (NCT04334629)	[70,71]
<i>Imatinib</i>	tyrosine kinase inhibitor; has been approved for treatment of many hematologic and solid neoplasm; as a weak base may be cumulated in lysosomes and lead to some antiviral effects during alkalization of lysosomes required for virus/cell fusion; has activity against <i>SARS-CoV</i> (<i>in vitro</i>) and <i>MERS-CoV</i> , and demonstrates also anti-inflammatory activity; used in combination with lopinavir/ritonavir, baricitinib and hydroxychloroquine (NCT04346147)	[72–75]
<i>Indomethacin</i>	non-steroidal anti-inflammatory drug; used to reduce fever, pain, stiffness, and swelling from inflammation; has a potent antiviral activity against <i>SARS</i> coronavirus	[76,77]
<i>Isotretinoin</i>	retinoid (synthetic analog of vitamin A); used to treat severe cases of acne vulgaris; isotretinoin has been reported to increase CD4 counts and significantly	[78,79]

	reduces viremia in <i>HIV</i> patients with acne vulgaris; it is suggested that isotretinoin therapy may affect the prevention of <i>SARS-COV-2</i> infection (NCT04361422)	
<i>Ivermectin</i>	anti-inflammatory drug, broad-spectrum anti-parasite medication; has also antiviral activity against a broad range of viruses; inhibits the replication of <i>SARS-CoV-2 in vitro</i> ; investigated also with hydroxychloroquine, nitazoxanide and favipiravir combination therapy	[80]
<i>L-Ascorbic acid</i>	vitamin C; mainly in intravenous use; may attenuate inflammation and vascular injury associated with sepsis and acute respiratory distress syndrome (ARDS); may also be effective in the treatment of COVID-19 pneumonia (NCT04323514)	[81–83]
<i>Leronlimab (PRO 140)</i>	monoclonal antibody, antiviral agent, belongs to a new class of HIV/AIDS therapeutics; used in CT of mild and critical <i>SARS-COV-2</i> infected patients	[84]
<i>Levofloxacin</i>	fluoroquinolone antibacterial agent; broad spectrum of activity against gram-positive and gram-negative bacteria also atypical respiratory pathogens (NCT02735707)	[85]
<i>Linagliptin</i>	DPP4 inhibitor; used to treat type 2 diabetes	[86,87]
<i>Losartan</i>	antihypertensive drug; angiotensin II receptor blocker; used to treat hypertension and renal disease; the main goal of one from many studies of losartan is identification whether or not ARBs have an impact on inhibiting the progression to respiratory failure in COVID-19 among patients with hypoxia (NCT04340557)	[26]
<i>Mavrilimumab</i>	human monoclonal antibody; investigated for the treatment of rheumatoid arthritis	[88,89]
<i>Methylprednisolone</i>	glucocorticosteroid; a drug with long-term anti-inflammatory and immunosuppressive effects; used in combination therapy with e.g. tacrolimus (NCT04341038) or siltuximab (NCT04329650)	[46,90]
<i>Naproxen</i>	non-steroidal, anti-inflammatory drug; inhibitor of both cyclooxygenase (COX-2) and of <i>influenza A</i> virus nucleoprotein (NP); study suggest that naproxen could combine a broad-spectrum antiviral activity with its well-known anti-inflammatory action that could help in reducing severe respiratory mortality associated with COVID-19	[91]
<i>Nintedanib</i>	inhibitor of tyrosine kinases; used in pulmonary fibrosis, systemic sclerosis-associated interstitial lung disease, and non-small cell lung cancer (NSCLC); actually efficacy and safety in the treatment of pulmonary fibrosis in patients with moderate to severe COVID-19 (NCT04338802)	[92,93]
<i>Nitazoxanide</i>	thiazolide compound with antiparasitic, antibacterial and antiviral activity; inhibits a broad range of influenza A and B viruses, replication of a wide variety of other RNA and DNA viruses (e.g. respiratory syncytial virus, parainfluenza, coronavirus, rotavirus, norovirus, hepatitis B, hepatitis C, dengue, yellow fever) in cell culture assays; in CTs study in mono or combined therapy with e.g. ivermectin (NCT04351347, NCT04341493), hydroxychloroquine (NCT04341493) or azithromycin (NCT04382846)	[94–96]
<i>Nitric Oxide</i> inhaled nitric oxide gas (NO)	used for the relaxation of smooth muscle cells in the vasculature; has shown to have antiviral activity against other coronavirus strains (e.g. during the 2003 <i>SARS</i> outbreak); greatly increased the survival rate of infected eukaryotic cells, suggesting its direct antiviral effect. Currently, high doses of inhaled NO against <i>SARS-CoV-2</i> are studied (e.g. NCT04338828)	[97–99]
<i>Nivolumab</i>	monoclonal antibody; usually used to treat melanoma and many types of cancer, e.g. lung, kidney head and neck, urothelial, colon and liver. The purpose of clinical study (NCT04343144) is to show the efficacy of nivolumab to treat patients with COVID-19 (in combination with standard treatments)	[100]
<i>Osetamivir</i>	antiviral drug; neuraminidase inhibitor; shows efficacy in the treatment of natural acquired influenza in humans, and might also protect people and animals from influenza; in over 15 CTs with different combination of drugs, e.g. hydroxychloroquine and azithromycin (NCT04338698), lopinavir/ritonavir (NCT04255017), favipiravir (NCT04349241), antibody-rich plasma (NCT04348877) or arbidol (NCT04255017)	[101,102]
<i>Peginterferon lambda 1a</i>	type III interferon (IFN); stimulates immune response; high therapeutic potential in COVID-19 – λ interferon is effective against SARS coronavirus (genetic	[103,104]

	similarities between <i>SARS-CoV</i> and <i>SARS-CoV-2</i> are evident, NCT04344600)	
<i>Pembrolizumab</i>	monoclonal antibody; used to treat different types of cancer, e.g. lung or head and neck	[105–107]
<i>Piclidenoson</i>	adenosine A3 receptor inhibitor; anti-inflammatory agent; tested for plaque-type psoriasis	[108,109]
<i>Piperacillin</i>	beta-lactam antibiotic; in beta-lactamase inhibitor combination; broad spectrum of antibacterial activity also in pneumonia	[110]
<i>Povidone-Iodine</i>	antiseptic; used for hand disinfection, skin preparation and antiseptic irrigation; study hypothesis: oral gargles and nasal rinses using a povidone iodine solution will help to reduce the viral load in the nasopharynx and oropharynx	[111–114]
<i>Prednisone</i>	Corticosteroide ;anti-inflammatory and immunosuppressive agent; in COVID-19 treatment, the reducing influence on the risk of acute respiratory distress syndrome is tested (NCT04344288)	[115]
<i>Remdesivir (GS-5734™)</i>	nucleotide analog; broad-spectrum antiviral drug; currently available evidence about effects against coronaviruses are favorable (<i>in vivo</i> and <i>in vitro</i> study); 35 CTs – mono and combined therapy with e.g. hydroxychloroquine, lopinavir/ritonavir (NCT04330690), tocilizumab (NCT04409262) or baricitinib (NCT04401579)	[116,117]
<i>Ribavirin</i>	guanosine nucleoside; broad-spectrum virustatic activity; actually in 9 CTs, three completed, one of them in combination therapy with lopinavir/ritonavir and IFN-β (NCT04276688)	[118–121]
<i>Ritonavir</i>	HIV protease inhibitor; in combination with other HIV therapies and antivirals, a potential treatment for COVID-19 caused by <i>SARS-CoV-2</i> , e.g. with xiyanning, (NCT04295551), danoprevir, (NCT04345276), ASC09 (NCT04261907) or interferon-β 1 and interferon-β 1b (NCT04343768); INF-β is even 10 times more effective than any other types of interferons tested against SARS and the most efficient antiviral drug against <i>SARS-CoV</i> so far), at the moment 80 CTs are performed in conjunction with e.g. danoprevir, lopinavir, hydroxychloroquine sulfate, interferon nebulization	[122–130]
<i>Rivaroxaban</i>	anticoagulant; direct factor Xa inhibitor; used for the prevention of venous thromboembolism in adults e.g. after knee replacement surgery or used to treat deep vein thrombosis and pulmonary embolism; is studied in patients with COVID-19 as an anticoagulant agent (NCT04416048)	[131,132]
<i>Ruxolitinib</i>	JAK1 and JAK2 selective inhibitor using in primary marrow fibrosis; responsible for multiple cellular signals including the proinflammatory interleukin-6 (IL-6); study hypothesis: stop of the dysregulated immune response caused by COVID-19 (NCT04334044)	[14]
<i>Sargramostim</i>	immunostimulator; stimulates white blood cell (WBC) production, and in particular, neutrophil, macrophage, and dendritic cell production; in CT the treatment in acute hypoxic respiratory failure due to COVID-19 is tested (NCT04326920)	[133]
<i>Sarilumab</i>	monoclonal antibody; inhibits the IL-6 receptor; and thereby may prevent of the overactive inflammatory response in the lungs of patients who are in severe or serious condition of COVID-19	[134]
<i>Sildenafil citrate</i>	PDE5 inhibitor; dilates blood vessels; COVID-19 study hypothesis: sildenafil citrate may help open the tiny vessels that draw oxygen from the lungs, allowing patients to overcome the respiratory distress that occurs in some cases of COVID-19 (NCT04304313)	[135]
<i>Siltuximab</i>	monoclonal antibody; anticancer drug, can be used as a treatment against prostate or metastatic renal cells or cancer; the use of this biopharmaceutical at the time of hyperinflammation episodes start, may improve the prognosis of the patients (NCT04329650)	[136]
<i>Simvastatin</i>	statin; used to lower the risk of cardiovascular disease and manage abnormal lipid levels; have anti-inflammatory and immunomodulatory effects	[137]
<i>Sirolimus</i>	mammalian target of rapamycin inhibitor; potent immunosuppressant and possesses both antifungal and antineoplastic properties	[138]
<i>T89 (Dantonic)</i>	botanical product, is a well-known traditional Chinese medicine; used for the treatment of e.g. angina pectoris (AP), myocardial infarction	[139]

<i>Tacrolimus</i>	immunosuppressor agent; used after organ transplant, in severe refractory uveitis, and the skin condition vitiligo	[140]
<i>Telmisartan</i>	angiotensin II type 1 receptor blocker; used to treat high blood pressure in monotherapy or in combination with other antihypertensive agents. Telmisartan has been proposed as an alternative treatment for COVID-19 patients prior to the development of ARDS (NCT04355936)	[141–143]
<i>Thalidomide</i>	anti-inflammatory, anti-fibrotic, anti-angiogenesis drug; effective in severe lung injury (e.g. against <i>H1N1</i> influenza); also used for the treatment of inflammation associated with Hansen's disease (leprosy) and as a chemotherapeutic agent for patients with multiple myeloma, purposes for which it was originally prescribed off-label	[144–146]
<i>Tocilizumab</i>	humanized monoclonal anti– interleukin 6 receptor antibody; it is hypothesized that it can reduce mortality in patients with severe COVID-19 prone to Cytokine-Release Syndrome (CRS) and ARDS Acute Respiratory Distress Syndrome (ARDS) (NCT04335071)	[147,148]
<i>Tofacitinib</i>	Janus kinase inhibitor; used to treat rheumatoid arthritis, psoriatic arthritis, ankylosing spondylitis, administration of tofacitinib in patients with symptomatic pneumonia pulmonary flogosisreductionis analyzed (NCT04332042)	[149–152]
<i>Tradipitant</i>	neurokinin 1 antagonist; has been used in trials studying the treatment and prevention of eczema, pruritus, gastroparesis, chronic pruritus, and atopic dermatitis	[153]
<i>Tranexamic Acid (TXA)</i>	blood clot stabilizer; used routinely for reduction of blood loss in surgery and trauma; inhibits conversion of plasminogen to plasmin and is proposed to be used against COVID-19 – plasmin, endogenous protease, acts on <i>SARS-CoV-2</i> virus by cleaving a newly inserted furin site in the S protein of the virus thus increasing infectivity and virulence (NCT04338074)	[154]
<i>Umifenovir (Arbidol)</i>	indole-based antiviral drug, dual-acting; used in prophylaxis and for the treatment of influenza and other respiratory infections; <i>in vitro</i> study have shown effect against coronaviruses	[155–157]
<i>Valsartan</i>	cardiovascular drug; AT1 angiotensin II receptor inhibitor; used to treat high blood pressure, cardiac infraction, heart failure, and diabetic kidney disease; may prevent the development of ARDS and avert morbidity and mortality (NCT04335786)	[158,159]

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