**Supplementary materials**

**Impact of Textile Dyes on Health and Ecosystem: A Review of Structure, Causes and Potential Solutions**

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**Supplementary tables:**

**Table S1.** Classification of dyes based on textile usage, chemical constitution and solubility (Broadbent 2001; Samsami et al. 2020)

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| **Classification** | | | **Types** | **Refs.** |
| Based textile usage | | | * Azoic dyes * Acid dyes * Basic dyes * Direct dyes * Disperse dyes * Mordant dyes * Pigments * Reactive dyes * Sulfur dyes * Vat dyes | (Broadbent 2001) |
| Based on chemical constitution | | | * Azo dyes * Anthraquinone dyes * Heterocyclic dyes * Indigoid dyes * Nitro dyes * Phthalocyanine dyes * Polymethine dyes * Stilbene dyes * Sulfur dyes * Triphenylmethane dyes |
| Based on solubility | Soluble | Anionic | * Acid dyes * Direct dyes * Mordant dyes * Reactive dyes | (Samsami et al. 2020) |
| Cationic | * Basic dyes |
| Insoluble | | * Azo dyes * Disperse dyes * Mordant dyes * Sulfur dyes * Vat dyes |

**Table S2.** The industrial dyes according to their application class, characteristics and application with structure.

|  |  |  |
| --- | --- | --- |
| Class of dye | Structure | Characteristics and application |
| Acid | C.I.Acid Red 35,C.I.18065,CAS 6441-93-6,523.45,C19H15N3Na2O8S2,Acid Red 3B,Acid Red 6B,Acid pink 3B,Acid Brilliant Red 3BSAcid Red 35 | **Characteristic:** The size of the dye molecule has a direct relationship with color fastness (Walters, A., Santillo, D., & Johnston 2005) |
| **Applications:** Acid dyes are used on a variety of textile substrates: wool, silk, paper, inks, leader, polyamide, cosmetics, ink-jet printing etc. |
| Reactive | https://www.researchgate.net/profile/Ramiro-Escalera/publication/318283587/figure/fig2/AS:513698172030976@1499486597026/Chemical-structure-of-the-dye-Reactive-blue-19_W640.jpgReactive Blue 19 | **Characteristics:**  Dyeing with reactive dye takes less time and is done at a lower temperature (above 60°) (Chiou and Li 2002; Pei et al. 2017). This dyes are now in powder, liquid, and print paste forms and are water soluble. |
| **Applications:** For dyeing cellulose, protein, and polyamide fibers, reactive dyes are utilized (Mathur et al. 2012). |
| Disperse | C.I.Disperse Blue 7,C.I.62500,CAS 3179-90-6,C18H18N2O6,358.35,Cibacet Turquoise G Disperse Blue 7 | **Characteristics:** Disperse dyes are comparatively low molecular weight (range; 400-600), slightly water soluble and substantive to hydrophobic fibers (Nylon & Polyester) (Ding et al. 2020). |
| **Applications:** The only dyes that may be used to dye poly (ethylene terephthalate) are disperse dyes. Furthermore, cellulose acetate, nylon, and polyester fibers are dyed with disperse dyes. |
| Direct | http://www.worlddyevariety.com/wp-content/uploads/2012/07/Direct-Orange-34.gifDirect Orange 36 | **Characteristic:** Direct dyes are less expensive than indirect dyes (Zinatloo-Ajabshir et al. 2017). |
| **Applications:** Cotton, viscose, paper, leather, and cellulose fibers are dyed with direct dyes. |
| Basic | C.I.Basic Blue 6,C.I.51175,CAS 966-62-1,310.78,C18H15ClN2O,Meldola's BlueCI Basic Blue 6 | **Characteristics:** Thebasic dyes are less expensive, not easily soluble in water but soluble in alcohol and methyted spirit (2015). |
| **Applications:** Basic dyes are used in paper, inks and synthetic fibers. |
| Vat | C.I.Vat Blue 20,C.I.59800,CAS 116-71-2,456.49,C34H16O2,Vat Navy Blue BO,Cibanon Blue BOA-OlVat Blue 20 | **Characteristics:** Vat dyes are water soluble, the reduced dyes has substantivity to cellulose, exhibit excellent wet and light fastness and after dyeing soluble dyes are oxidized with in the fabric to form insoluble again(Khatri et al. 2015). |
| **Applications:** Vat dyes are used in viscose, wool, cotton and cellulose. Besides, this dyes are used in superior quality shirting material, military uniforms, furnishing, toweling and denim. |
| Sulfur | CAS No.1327-73-7,Sulphur Green 3 SuppliersSulfur brilliant green, CI 53570 | **Characteristics:** Sulfur dyes are water insoluble, it needs solubilization for application, and this dyes are used in alkaline condition. Additionally, electrolyte can be injected to the dye to enable faster the dye exhaustion process, which is optimal for generating black and brown on textile materials at a temperature of 90° (Chakraborty and Jaruhar 2014). |
| **Applications:** Sulfur dyes are mainly applied on cotton, viscose and staple fibers (Nguyen and Juang 2013). |
| Azoic | Disperse Yellow 3 Dye content 30 % | 2832-40-8Disperse Yellow 3 | **Characteristics:** Azoic dyesare water insoluble, They aren't dyes that have been pre-mixed. The color is generated in the fiber by two main components typically known as "Napthols" and "Bases". The dyed goods exhibit good to excellent light fastness and good washing fastness (Walters, A., Santillo, D., & Johnston 2005; Hassan and Carr 2018) |
| **Applications:** Azoic dyes are used in textile fibers such as cellulose acetate, polyester, rayon and cotton. |
| Mordant | Mordant Black 17Mordant Blue 17 | **Characteristics:** Many standard mordant dyes form stable complex on the Nano crystalline surface TiO2 (Millington et al. 2007)**,** some types of textile substrates have a poor affinity for them. These dyes are also anticipating a reaction from their industrial applications (2009). |
| **Applications:** Textile fibers such as silk, leather, and wool are dyed with mordant dyes. |
|  | Disperse Red 82Disperse Red 82 | **Characteristic:** Disperse azo dyes have a lower water solubility (Vacchi et al. 2016). |
| **Applications:** This dyes are used in polyester, polyamide and plastic. |

**Table S3.** Chemical class distribution across major application ranges (%) (Suteu, Zaharia and Malutan, 2012; R Ananthashankar, 2013).

|  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| **Chemical class** | **Distribution between application ranges, (%)** | | | | | | | | |
| **Acid** | **Basic** | **Direct** | **Disperse** | **Mordant** | **Pigment** | **Reactive** | **Solvent** | **Vat** |
| Unmetallised azo | 21 | 4 | 30 | 12 | 11 | 7 | 9 | 6 | - |
| Metal complex | 64 | - | 11 | - | - | - | 13 | 12 | - |
| Thiazole | - | 6 | 94 | - | - | - | - | - | - |
| Stilbene | - | 3 | 97 | - | - | - | - | - | - |
| Anthraquinone | 14 | 3 | - | 26 | 2 | 4 | 5 | 10 | 36 |
| Indigoid | 2 | - | - | - | - | 18 | - | - | 80 |
| Quinophthalene | 30 | 20 | - | 40 | - | - | 10 | - | - |
| Aminoketone | 10 | - | - | 40 | 8 | - | 3 | 8 | 21 |
| Phtalocyanine | 15 | 3 | 8 | - | 4 | 10 | 42 | 16 | 2 |
| Formazan | 69 | - | - | - | - | - | 31 | - | - |
| Methine | - | 70 | - | 24 | - | 2 | - | 4 | - |
| Nitro, nitroso | 30 | 3 | - | 48 | 3 | 4 | - | 12 | - |
| Triarylmethane | 36 | 21 | 1 | 1 | 24 | 4 | - | 13 | - |
| Xanthene | 32 | 16 | - | - | 10 | 2 | 2 | 38 | - |
| Acridine | - | 92 | - | 4 | - | - | - | 4 | - |
| Azine | 40 | 40 | - | - | - | 2 | - | 18 | - |
| Oxazine | - | 23 | 16 | 2 | 39 | 10 | 10 | - | - |
| **Thiazine** | **-** | 56 | - | **-** | 10 | - | - | 10 | 24 |

**Table S4.** Different dye classes have varying rates of fixation on different textile substrates (Dos Santos, Cervantes and Van Lier, 2003; Scalbi, Tarantini and Mattioli, 2005; Avvannavar, Mani and Kumar, 2008; Mani, Chowdhary and Bharagava, 2019).

|  |  |  |  |
| --- | --- | --- | --- |
| **Dye class** | **Fiber type** | **Fixation rate (%)** | **Effluent rate (%)** |
| Acid | Polyamide | 90-95 | 5-10 |
| Azo | Cellulose | 90-95 | 5-10 |
| Basic | Acrylic | 90-100 | 0-10 |
| Direct | Cellulose | 75-95 | 5-25 |
| Disperse | Polyester | 95-100 | 0-5 |
| Metal complex | Wool | 95-98 | 2-5 |
| Reactive | Cellulose | 50-90 | 10-50 |
| Sulfur | Cellulose | 65-90 | 10-35 |
| Dye-stuff | Cellulose | 85-95 | 5-15 |
| Vat | Cellulose | 80-95 | 5-20 |

**Table S5.** A typical textile industry effluent in terms of physio-chemical characteristics

|  |  |  |
| --- | --- | --- |
| Factor/ Parameter | Reported value\* | References |
| BOD | 237.2±32.1 | (Kaur et al. 2018; Tara et al. 2019; Hussain et al. 2019; Chandanshive et al. 2020) |
| TDS | 8850±756 | (Khan and Malik 2014; Chandanshive et al. 2017; Kaur et al. 2018; Kadam et al. 2018) |
| COD | 1268±121 | (Chandanshive et al. 2017; Kaur et al. 2018; Kadam et al. 2018; EL-Mekkawi et al. 2020) |
| PH | 8.75±1.29 | (Hussain et al. 2019; Oktem et al. 2019) |
| TSS | 253.2±43.5 | (Guadie et al. 2017; Tara et al. 2019; Hussain et al. 2019; EL-Mekkawi et al. 2020) |
| TS | 5076±344 | (Tara et al. 2019; Hussain et al. 2019) |
| EC | 7.1±1.72 | (Tara et al. 2019; Hussain et al. 2019) |
| TOC | 222.2±53.3 | (Tara et al. 2019; Hussain et al. 2019; Ağtaş et al. 2021) |
| Cl- | 51.6±16 | (Tomei et al. 2016; Guadie et al. 2017) |
| Cr | 2.74±0.4 | (Chandanshive et al. 2017, 2020; Watharkar et al. 2018; Hubadillah et al. 2020) |
| Pb | 0.35±0.3 | (Amare et al. 2017; Chandanshive et al. 2017, 2020; Hubadillah et al. 2020) |
| AIk | 396±132 | (Guadie et al. 2017; Arcanjo et al. 2018) |
| SO42- | 240.6±75.4 | (Arcanjo et al. 2018; Hussain et al. 2019) |
| TN | 24.4±11.7 | (Guadie et al. 2017; Tara et al. 2019; Hussain et al. 2019; EL-Mekkawi et al. 2020) |
| Phenol | 0.52±0.22 | (Tara et al. 2019; Hussain et al. 2019) |
| As | 2.21±0.4 | (Chandanshive et al. 2017, 2020; Watharkar et al. 2018; Kadam et al. 2018) |
| Zn | 0.37±0.37 | (Amare et al. 2017; Hubadillah et al. 2020) |
| NO3- | 116.1±109 | (Arcanjo et al. 2018) |
| Cu | 0.54±0.5 | (Amare et al. 2017; Hubadillah et al. 2020) |
| PO43- | 12.4±3.2 | (Guadie et al. 2017; Tara et al. 2019; Hussain et al. 2019; EL-Mekkawi et al. 2020) |
| Cd | 0.62±0.3 | (Tara et al. 2019; Hussain et al. 2019; Hubadillah et al. 2020; Chandanshive et al. 2020) |

*Note: All the values are means of triplicates (n = 3) ± SD.*

*\*Except pH, all the parameters are expressed in “mgL-1”, but the conductivity is expressed in “μmho/cm”.*

**Supplementary figures:**

Figure S1.Major sectors that discharge dyes into the environment. Reproduced with permission (Samsami et al. 2020). Copyright 2020, Elsevier.

**Figure S2.** International textile dyes market over the forecasted period of 2016–2023. Reproduced with permission (Samsami et al. 2020). Copyright 2020, Elsevier.

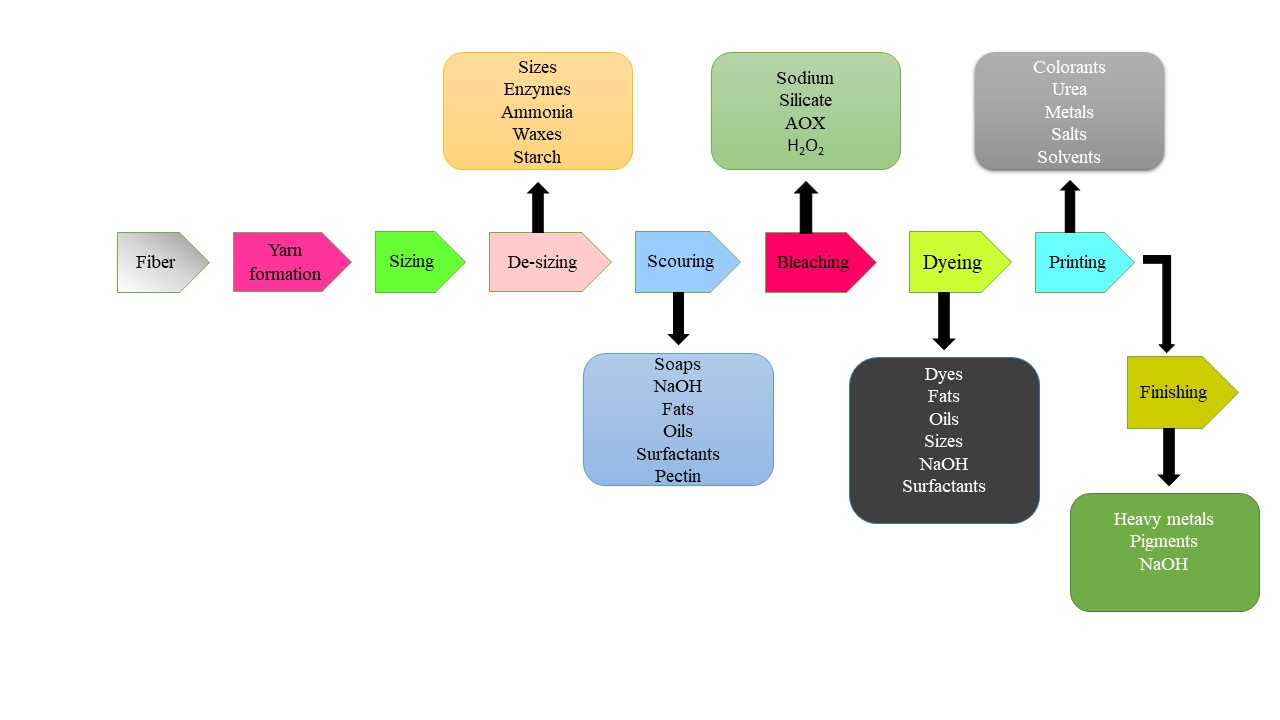


Figure S3. Different wet processing steps in the textile industry and some pollutants propagated from these steps.

|  |  |  |
| --- | --- | --- |
| A black and white striped caterpillar on a mossy rock  Description automatically generated with low confidence | A picture containing pan  Description automatically generated | A picture containing outdoor, grass, outdoor object  Description automatically generated |

**Figure S4.** Discharge colored dyes from textile industries.

Diagram

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**Figure S5.** Sources and pathways of dyes in the eco-system (Dutta et al. 2021).

Diagram

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**Figure S6.** Manufacturing processes in textile industry, wastewater discharge, negative impacts and several remedial techniques. Reproduced with permission (Kishor et al. 2021). Copyright 2021, Elsevier.

Diagram

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**Figure S7.** Industrial dye effluent techniques (Hynes et al. 2020).

**Diagram

Description automatically generatedFigure S8.** Suggested mechanism for reduction of azo dyes by azo reductase. Reproduced with permission (Pearce et al. 2003). Copyright 2003, Elsevier.

Diagram

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**Figure S9.** schematic illustration of coagulation-flocculation for wastewater treatment (B) (Choumane et al. 2017).

**Diagram

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Diagram

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**Figure S11.** Process in the removal of dyes using white rot fungi (Jebapriya and Gnanadoss 2013).

**Diagram

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**Figure S12.** The azo dye (AO7) degradation pathway by the oleaginous yeast consortium NYC-1 linked to biodiesel synthesis. (Ali et al. 2021).

Diagram

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**Figure S13.** GC–MS chromatograms of: (a) raw wastewater, (b) treated by microbubble-ozonation, and (c) treated by macrobubble-ozonation. Reproduced with permission (Zheng et al. 2015). Copyright 2015, Elsevier.

Diagram

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**Figure S14.** Broad overview and classification of different AOPs, influencing factors and published EEO-values of different AOPs sorted according to median values. Reproduced with permission (Miklos et al. 2018). Copyright 2018, Elsevier.

Diagram, engineering drawing

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**Figure S15.** Schematic diagram of the MF experimental apparatus. Reproduced with permission (Lu and Liu 2010). Copyright 2010, Elsevier.

Diagram, schematic

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**Figure S16.** Operational modes of the pilot scale reverse osmosis process. (A) Batch (or concentrated) mode of operation, (B) Complete recycle mode of operation, (C) Continuous mode of operation. Reproduced with permission (Sahinkaya et al. 2019). Copyright 2019, Elsevier.

**Diagram

Description automatically generated****Figure S17.** Types and benefits of different nanomaterials membrane bioreactor (NMs-MBR) technology and several publications are observed related to these NMs-MBR technology with their historical timeline. a. Examples of commonly used nanomaterials membrane bioreactor (NMs-MBR) technology (left to right) nanofibers membrane bioreactor (NFs-MBR), nanoparticles membrane bioreactor (NPs-MBR), nanotubes membrane bioreactor (NTs-MBR), nanocrystals membrane bioreactor (NCs-MBR), nanowires membrane bioreactor (NWs-MBR), nanosheets membrane bioreactor (NSs-MBR), and the advantages of using NMs-MBR technology are fouling control, high efficiency and sustainability. b. Diagram of the total number of publications related to different types of NMs-MBR technology. Until 3rd August 2020, which were collected from the web of science scientific database. c. Historical development of NMs-MBR technology for wastewater treatment. In 2005, Tae-Hyun Bae investigated the ability of TiO2-embedded nanocomposite membrane for NPs-MBR , In 2009, Decostere Bjorge evaluated the electrospun NFs-MBR , In 2014, Chuanqi Zhao prepared nanosheets membrane and tested for NSs-MBR , In 2015, Zahra Rahimi applied NTs-MBR , In 2018, Jinling Lv synthesized nanocrystal membrane and used for NCs-MBR and In 2019, Xiafei Yin established nanowires membrane and used for membrane bioreactor (NWs-MBR) (Pervez et al. 2020).

**Diagram, schematic

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**Figure S18.** Schematic diagrams of AFMBR (a) and anoxic-aerobic MBR (b). Reproduced with permission (Li et al. 2020)**.** Copyright 2020, Elsevier.

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