

**OCCUPATIONAL EXPOSURES IN
PAINT MANUFACTURE AND PAINTING**

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1. Historical Perspectives and Description of Painting Trades

1.1 Description of paint products

The term *organic coating* encompasses conventional paints, varnishes, enamels, lacquers, water-emulsion and solution finishes, nonaqueous dispersions (organosols), plastisols and powder coatings. The following definitions have been used commonly, although they have not always been strictly applied.

Paint is a suspension of finely divided pigment particles in a liquid composed of a binder (resin) and a volatile solvent, sometimes with additives to impart special characteristics. The volatile solvent evaporates from the drying film after application, while the binder holds the pigment in the dry film, causing it to adhere to the substrate. Some high quality, hard gloss paints are referred to as *enamels* (Piper, 1965; Schurr, 1981).

Lacquer is defined as a coating that dries primarily by evaporation rather than by oxidation or polymerization. Because the solvents used in lacquers are relatively volatile and no chemical change is required for formation of the film, lacquers dry very rapidly (Piper, 1965; Hamilton & Early, 1972).

Varnish is defined as a homogeneous, transparent or translucent liquid that is converted to a solid, transparent film after being applied as a thin layer (Schurr, 1981).

The basic components of paints may have a widely varying chemical composition, depending on the colour, durability and other properties required from the paint.

(a) *Pigments*

Pigments can be classified as (i) inorganic, (ii) organic and (iii) earth pigments, such as ochre. They can also be classified into whites, colours, metallic flake pigments and powders. They are generally added in considerable proportion by weight (20–60%) and are used in paints to provide colour, opacity and sheen and also affect the viscosity, flow, toughness, durability and other physical properties of the coating. The physical properties of pigments, such as particle shape and size, vary; the diameter of the particles is generally $< 3 \mu\text{m}$. The particles in dry pigment powders (0.5–10 μm) are partially in the range of respirable dust (Krivanek, 1982).

(i) *Inorganic pigments*

Inorganic pigments are an integral part of numerous decorative, protective and functional coating systems, such as automobile finishes, marine paints, industrial coatings, traffic paints, maintenance paints, and exterior and interior oil, alkyd and latex house paints. Inorganic pigments belong to numerous chemical classes, primarily including elements, oxides, carbonates, chromates, phosphates, sulfides and silicates (Schiek, 1982).

Many forms of lead have been used for more than 200 years in pigments; these include carbonate (white lead), oxides (litharge, red lead), sulfate, oxychloride (Turner's yellow), acetate, borate and chromates (IARC, 1980a,b; Schiek, 1982).

Zinc chromate, little known before 1914, was widely employed during the Second World War to inhibit rust on all sorts of equipment (Brunner, 1978). Other chromium pigments that have been used in paint for many years include lead chromates, barium chromate and chromium oxide (IARC, 1980b). Other inorganic pigments include cadmium sulfide, cadmium sulfoselenide and antimony trioxide (see monograph, p. 291). Various grades of naturally occurring ferric oxide provide yellow, red and brown pigments (Schiek, 1982).

White pigments constitute over 90% of all pigments used (Krivanek, 1982). Until the nineteenth century, white lead in linseed oil was used primarily, and prior to 1920 the available white pigments were basic carbonate white lead, basic sulfate white lead, zinc oxide, leaded zinc oxide and lithopone (Martens, 1964; Federation of Societies for Paint Technology, 1973). Increasing awareness of the toxic hazards of white lead stimulated the development of other pigments, which became available to paint manufacturers in the early part of the twentieth century (Brunner, 1978).

The most common pigment employed in paint is the white pigment titanium dioxide (see monograph, p. 307), produced in two different crystal forms - rutile and anatase. Although it was introduced shortly after 1918, it was not used widely because of its high cost. The first titanium dioxide pigment was a composite of 30% titanium dioxide (anatase crystal structure) and 70% barium sulfate. A major gain was made by the production of titanium dioxide with the rutile crystal structure, which has almost 25% greater opacity than the anatase form. Because of the chemical inertness of titanium dioxide, its extreme whiteness, excellent covering power and lack of toxicity, compared to white lead, it soon dominated in the manufacture of white paint and, by 1945, represented 80% of white pigment on the market. Concomitantly, the use of white lead in paints fell during 1900-45 from nearly 100% to less than 10%. The share of lithopone, a coprecipitate of 28-30% zinc sulfide and 70-72% barium sulfate (Schiek, 1982), introduced before the First World War, rose to 60% by about 1928 but fell to 15% by 1945 (Brunner, 1978). Calcium carbonate and aluminium silicate have also been used as white pigments.

The most common metallic dusts and powders used in paint are aluminium powder, zinc dust (Schiek, 1982) and bronze powders, which consist of metals in a finely divided state; e.g., gold bronzes are alloys of copper with varying proportions of zinc or aluminium.

Materials used as extender pigments include barium sulfate (barytes), calcium carbonate (ground limestone and chalk), silica (diatomaceous and amorphous; see IARC, 1987a,b), clays (hydrated aluminium silicate), talc (hydrated magnesium silicate; see IARC, 1978c,d) and mica (hydrated potassium aluminium silicate). These minerals are often added to paint to reduce cost, improve physical characteristics and increase resistance to wear; their effects are largely governed by the average particle size (Martens, 1964).

(ii) *Organic pigments*

Hundreds of organic pigments, comprising a broad spectrum of structural classes, are used in the paint industry. Organic pigments may be classified as azo or nonazo pigments.

Azo pigments are formed by successive diazotization of a primary amine and coupling. Monoazo and diazo pigments contain, respectively, one and more than one chromophore ($-N=N-$) group and are subdivided into two types, the pigment dyes and the precipitated azo dyes. The most important and established uses for pigment products include the coloration of surface-coating compositions for interior, exterior, trade and automotive applications, including oil and water emulsion paints and lacquers (Fytelson, 1982).

Prior to the discovery of Perkins' mauve in 1856, colour was obtained from natural sources, i.e., woad, madder, indigo, cochineal and log wood. The development of synthetic colouring materials continued with the discovery of fuchsin in 1858 and of other triphenylmethane dyes, such as alkali blue, methyl violet and malachite green. Lakes of these dyes were used as the first synthetic organic pigments. The largest single advance in pigment technology after the First World War was the discovery in the 1930s of phthalocyanine blue (Monastral blue) and, later, its halogenated derivatives (Monastral greens) which were widely used in automotive finishes (Brunner, 1978; Fytelson, 1982). Other main categories of nonazo organic dyes and pigments used in paints and related products include quinacridones, thioindigos, perinones, perylenes and anthraquinone (Fytelson, 1982).

(iii) *Earth pigments*

Iron oxides are the most widely used of the coloured pigments derived from natural sources. Natural iron oxides are processed from several different ores, including haematite (see IARC, 1972, 1987e), limonite, siderite and magnetite, and provide a range of reds, yellows, purples, browns and blacks (Schiek, 1982).

(b) *Binders (resins)*

The vehicle portion of paints contains components collectively termed 'binders', which hold the pigment in the dry film and cause it to adhere to the surface to be painted. Almost all binders in modern paint films are composed of polymer materials, such as resins, and drying oils, whose main functions are to provide film hardness, gloss, surface adhesion and resistance of the film to acids, alkalis and other agents (Krivanek, 1982). A large variety of both natural and synthetic resins has been employed in paints. Natural resins have been used in paints for centuries, while synthetic resins have been commercially available since the early 1900s.

(i) *Natural resins and oils*

From early times, various natural resins have been used to reinforce linseed oil and other drying oils, since paints based only on pigment and oil yield only very soft films. Shellac (Brunner, 1978) and insect exudations are natural oleoresins that have been used in paints for centuries. Oleoresins from tree saps (shellac) are a mixture of single- and fused-ring compounds with various oxygenated groups possessing a wide range of molecular weights, solubilities and chemical and physical properties.

Kauri, a fossil resin, was used widely but had been replaced by the beginning of the twentieth century by Congo copal, which is a much harder resin but which requires prolonged heat treatment, known as 'gum running'. The term 'copal' is a generic name covering

a number of fossilized and recent resins found in many tropical and subtropical parts of the world, which include all the harder resins used for oil varnishes (Brunner, 1978).

Another useful natural resin is rosin (colophony), which is obtained as a residue after distilling pine oleoresin for the production of turpentine. Rosin consists of about 85% rosin acids and 15% neutral substances and can be classified into two main types – gum rosin and wood rosin. Rosin has been used in paints (principally alkyd resins) for many years. They are often upgraded to yield higher quality resins by chemical reactions, including liming (calcium rosinate), esterification of rosin with glycerol, and reactions with trimethylolpropane, phthalic anhydride, maleic anhydride, adipic acid and sebacic acid (Krivanek, 1982).

Vegetable and fish oils have long been used as binders in traditional paints and varnishes. White linseed oil has been the most important oil in standard exterior paints, despite its moderately slow drying rate. It is infrequently employed in interior paints because of yellowing. Other important oils include castor oil, tall oil, soya bean oil, coconut oil, cottonseed oil, tung oil and various fish oils (Brunner, 1978; Lowell, 1984).

Although raw oils are useful as paint binders, it has been advantageous to use them in conjunction with refined oils and oils treated with heat to increase viscosity (heat-bodied oils) which isomerize the oil and improve the drying rate of the films. Oleoresinous varnishes are made by cooking oils with natural or synthetic resin, resulting in more rapid drying and a harder film (Lanson, 1978).

(ii) *Synthetic resins*

A wide variety of synthetic resins has been commercially available since the early 1900s. Those that have been most frequently employed in paints, varnishes and lacquers include cellulosic, phenolic, alkyd, vinyl, acrylic and methacrylic, and polyurethane resins, chlorinated rubber derivatives, styrene-butadiene and silicone oils (Martens, 1964; Krivanek, 1982).

Mixtures of different synthetic resins are often incorporated into a paint to furnish certain properties not provided by a single resin. While the amount of resin in paint varies, values of 20–35 wt% are common (Krivanek, 1982). The choice of a resin(s) for a particular application depends on factors such as appearance, ease of application, cost and resistance to chemicals, heat and wear.

Some resins (polyurethanes, epoxys; see IARC, 1976a) are blended immediately before use with cross-linking agents between the individual polymer chains, resulting in a hard, serviceable film.

Alkyd, acrylic, polyurethane and polyester resins have broad areas of use in paints, including paints for houses, automobiles, furniture and appliances, as well as in the protection of metal surfaces, e.g., in chemical plants and oil refineries.

Phenolic resins: The first synthetic resins used in paints were phenolic resins, which were introduced in the 1920s and are made from formaldehyde (see IARC, 1982a, 1987f) and phenol (see monograph, p. 263) or substituted phenols in the presence of alkaline or acid catalysts. Depending on the type and proportion of reactants, and on the reaction conditions, the resins may be heat-reactive or not. The first product of the reaction is methylol phenol, which reacts further. With an excess of formaldehyde under alkaline conditions,

methylol groups react slowly with phenol, are retained in the reaction product and can act as reactive sites in varnish preparations or for cross-linking in finished products (Lowell, 1984).

The early phenolic resins developed between 1905 and 1910 were based on unsubstituted phenols, e.g., cresols, *para*-phenylphenol and *para-tert*-butylphenol, which are oil-soluble, and constituted a new type of varnish with superior hardness and resistance to water, solvents, chemicals and heat. Heat-sensitive phenolic resins that are insoluble in oil may be dissolved in solvents and employed as the sole vehicle for metal coatings (Lowell, 1984).

Alkyd resins: The advent of alkyd resins is considered to be a major breakthrough in modern paint technology. Alkyds are oil-modified polyester resins produced by the condensation reaction of polyhydric alcohols, polybasic acids and monobasic fatty acids, e.g., linseed or soya fatty acids (Lowell, 1984). The specific definition, which has gained wide acceptance, is that alkyds are polyesters modified with monobasic fatty acids (Lanson, 1978). The alkyds used initially were principally products of the chemical reaction of phthalic anhydride and glycerol with certain vegetable oils or their corresponding fatty acids (Brunner, 1978).

In recent years, the terms 'non-oil' and 'oil-free' alkyd have been used to describe polyesters formed by the reaction of polybasic acids with polyhydric alcohols in excess of stoichiometric amounts. These products are best described as saturated polyesters containing unreacted -OH or -COOH groups (Lanson, 1978).

Monobasic acids modify the properties of alkyd resins by controlling functionality, and thus polymer growth, as well as by the nature of their inherent physical and chemical properties. The majority of monobasic acids used in alkyd resins are derived from natural glyceride oils and are in varying degrees of unsaturation. The most common fatty acids present in these oils include lauric, palmitic, stearic, oleic, linoleic, linolenic, eleostearic, ricinoleic and licanic acids. Alkyd resins with relatively high fatty acid contents are called 'long-oil' alkyds; when the oil percentage is relatively low, they are known as 'short-oil' alkyds (Lanson, 1978).

Although glycerol and pentaerythritol are the major polyhydric alcohol components of alkyd resins, a number of other polyols are employed to a lesser degree, including sorbitol, trimethylolethanol, trimethylol propane, dipentaerythritol, tripentaerythritol, neopentylglycol and diethylene glycol. The principal polybasic acid for alkyd resins is phthalic acid, which is prepared and used as the anhydride. Isophthalic anhydride is also employed to yield somewhat faster drying and tougher, more flexible films than the analogous *ortho*-phthalic resins.

Long-oil alkyds are soluble in mineral spirits and are widely employed in architectural brushing enamels, exterior trim paints and wall paints, and their flexibility and durability have made them useful for top-side marine paints, metal maintenance paints and as a clear varnish. Medium-oil alkyds are the most versatile of the alkyd class, and their superior air drying, flexibility and durability allow for their use in maintenance paints, metal primers and a variety of general-purpose enamels. Short-oil alkyds are either drying or nondrying, require a strong aromatic solvent, such as toluene or xylene (see monographs, p. 79 and p. 125), and have been employed principally as industrial baking finishes.

Many polymeric materials and reactive functional materials can be used to produce suitably designed alkyds and to impart improved and/or special film-forming properties, in-

cluding nitrocellulose, polyisocyanates, urea-formaldehyde resins, silicones, melamine-formaldehyde resins, reactive monomers, phenolic resins, cellulose acetobutyrate, chlorinated rubber, phenolic varnishes, vinyl resins, polyamides, chlorinated paraffin, natural resins, epoxy resins and monobasic aromatic acids (Lanson, 1978).

Short-oil alkyd resins with a phthalic anhydride content of 38–45% contain a higher proportion of hydroxyl groups, which provide reactive sites for alkylated urea-formaldehyde and melamine-formaldehyde resins. These alkyds are generally based on tall-oil fatty acids, soya bean oil or fatty acids. Amino-alkyd resins are widely used in industrial baking enamels (Lanson, 1978).

The compatibility of alkyd resins with nitrocellulose extends up to 55% nitrocellulose content, and nitrocellulose lacquers are produced in large quantities. Alkyds modified with short-chain acids, such as those from coconut oil and castor oil, are widely used in high-grade furniture lacquers (Lanson, 1978).

The general effect of the alkyd modified resins has been to upgrade the gloss, adhesion and durability of nitrocellulose lacquers. Alkyd resins have been used in protective coatings for over 40 years, and they still rank as the most important synthetic coating resin, constituting about 35% of all resins used in organic coatings (Lanson, 1978). The largest market for alkyds in product finishes includes machines and equipment and wood and metal furniture and fixtures (Connolly *et al.*, 1986).

Vinyl resins: Vinyl polymers and copolymers were among the first synthetic polymers and are widely employed in trade paints. Although synthesis of polyvinyl chloride (see IARC, 1979a) was first reported in 1872 and that of polyvinyl acetate (see IARC, 1979b) in 1913, neither was developed commercially until the mid-1920s (Powell, 1972).

Vinyl monomers can be induced to polymerize readily by the addition of initiators, such as peroxides and azo compounds, which decompose at reactor temperature to generate free radicals. Polymerization processes involve radical formation, initiation, propagation, including chain transfer, and termination (Powell, 1972). The principal vinyl resins of importance in the paint industry are polyvinyl chloride, polyvinyl acetate and polyvinyl butyrate, which are available in a range of different compositions for specific uses and in grades that can be handled as true solutions in organic solvents, as high-solid dispersions ('organosols' or 'plastisols'), as dry powders or as water-borne latexes. Polyvinyl acetate is extensively used in emulsion paints, providing exceptional flexibility, toughness and water and chemical resistance. Vinyl chloride copolymer coatings are used in coil coatings and in industrial and marine coatings (Lowell, 1984; Connolly *et al.*, 1986).

Water emulsions of high molecular-weight polyvinyl acetate have been widely used in interior house paints. Copolymers of vinyl acetate with acrylic monomers are also employed in exterior emulsion house paints. Latexes of vinyl chloride polymers and copolymers have been commercially important for a number of years, e.g., as copolymers in exterior house paints, which often include a vinyl chloride-acrylic ester copolymer modified with a specially designed alkyd resin. Polyvinyl acetate and vinyl acetate copolymers are used in latex-based interior and exterior paints (Powell, 1972).

The principal modifying monomers that have been used with vinyl acetate include dibutyl maleate and fumarate, butyl-, 2-ethylhexyl- and isodecyl acrylates and higher vinyl esters (Powell, 1972). Copolymers of the acrylates and vinyl acetate are commonly called vinyl acrylics and generally contain 15% acrylic monomer by weight (Connolly *et al.*, 1986).

Acrylic and methacrylic ester resins: Acrylic resins have been divided into four specific types: water-based, solvent-based thermoplastic (lacquer types), solvent-based thermosetting and powder coating resins (Connolly *et al.*, 1986).

Commercial acrylic and methacrylic polymers are made from a variety of acrylic and methacrylic monomers (see IARC, 1979c). The major monomers used are the methyl, ethyl, butyl and 2-ethylhexyl esters of acrylic and methacrylic acids, which readily undergo polymerization in the presence of free-radical initiators, such as peroxides, to yield high molecular-weight polymers (Allyn, 1971; Lowell, 1984; Connolly *et al.*, 1986).

Monomeric acrylic esters are produced commercially by several processes based on ethylene cyanohydrin, acetylene, β -propiolactone (see IARC, 1974a) and ethylene oxide (see IARC, 1985a, 1987g). The acetone-cyanohydrin process is the major method for the production of monomeric methacrylate esters (Allyn, 1971).

Several types of functionality can be incorporated into acrylic and methacrylic monomers. These are principally the amide, carboxyl, hydroxyl and epoxy types and are used to confer cross-linking capabilities and thermosetting properties on the resulting polymers (Allyn, 1971). Other monomers are used in conjunction with the acrylic monomers to achieve different properties, including vinyl acetate (see IARC, 1986a), styrene (see IARC, 1979d), vinyl toluene, acrylonitrile (see IARC, 1979a, 1987h) and methylacrylamide (Allyn, 1971).

Acrylic and methacrylic polymers are used in the formulation of clear and pigmented lacquers. Dispersions in water and in organic solvents provide latex and organosol coatings, respectively. Polymethacrylates are harder and less flexible than the corresponding acrylates (Lowell, 1984).

Although thermoplastic acrylic emulsions have been commercially available since 1925, they were not widely used in coatings until 1953 when new grades specifically designed for paints were introduced (Allyn, 1971). In the late 1950s, lacquers of greatly improved durability, based on polymethylmethacrylate or thermosetting acrylic enamels were adopted by the automobile industry (Lowell, 1984). By the 1960s, the use of acrylic emulsion polymers had been firmly established in exterior coatings for wood surfaces, a field long dominated by oil paints (Allyn, 1971).

Epoxy resins: Epoxy resins were first derived from bisphenol A and epichlorohydrin (see IARC, 1976b, 1987i) and introduced into the paint industry in the late 1940s. Two major types of epoxy resin exist - glycidyl ether epoxy resins (see monograph, p. 237) and epoxidized olefins - the former of which is the most important. Epoxy resins based on bisphenol A and epichlorohydrin are the most prominent of the glycidyl ether category and are produced by a condensation reaction in which bisphenol A and epichlorohydrin are reacted in the presence of alkali (Allen, 1972). The resultant diglycidyl ether resin has a functionality of two reactive epoxy groups per molecule. Epoxy resins can be polymerized through their reactive epoxy group using amines or polyamides (Allen, 1972; Lowell, 1984).

Epoxy resins of a second major type, epoxidized olefins, are based on epoxidation of the carbon-carbon double bond. Coating compositions derived from epoxidized olefin have better weathering characteristics than analogous systems based on bisphenol A diglycidyl ether resins (Allen, 1972).

In order to proceed from the relatively low molecular weight of the coating composition, as applied, to the high molecular-weight polymer necessary for optimal film properties, a 'curing' or polymerization must take place, which can involve either the epoxide or free hydroxy groups in the resin, or a combination of the two. Some of the principal reactions that have been used include chemical cross-linking *via* the amine-epoxide reaction, an anhydride-epoxide reaction, reaction with methylol groups, e.g., between the secondary hydroxyl groups of the higher molecular-weight resins and the methylol groups of phenol-formaldehyde and urea-formaldehyde resins, cross-linking *via* the isocyanate-hydroxyl reaction and esterification reactions between solid-grade epoxy resins and carboxyl-containing compounds, particularly drying-oil fatty acids (Allen, 1972).

Solid-grade glycidyl ether resins are readily soluble in polar solvents, such as ketones, esters and ether-alcohols, as well as in chlorinated hydrocarbon solvents (Allen, 1972).

Glycidyl ether resins of high molecular weight (number average¹, about 7000; weight average², about 200 000) are unique among epoxy coatings in that they form coatings by solvent evaporation alone (Allen, 1972). Because of their toughness, adhesion and corrosion resistance, epoxy resins are used in many applications, including industrial maintenance, automobile primers and coatings for appliances and steel pipes. Epoxys combined with phenolic resins and thermosetting acrylic resins yield high bake finishes with hardness, flexibility and resistance to chemicals and solvents (Lowell, 1984).

Polyurethane resins: Although polyurethanes were synthesized in 1937, the utility of weather-resistant polyurethane coatings became manifest only in the 1960s. Polyurethanes are obtained from the reaction of polyhydric alcohols and isocyanates. Nonreactive polymers can be prepared by terminating the polymer chains with monofunctional isocyanates or alcohols. Cross-linked polymers are formed from polyfunctional isocyanates or alcohols (Lowell, 1984). Isocyanates that have been employed include toluene diisocyanate (see IARC, 1986b) and hexamethylenediisocyanate.

Because of the wide range of physical properties obtained through variations in formulating polyurethane coatings, they are used in industrial and maintenance coatings, as well as in coatings for wood, concrete and flexible structures (Lowell, 1984). Polyurethane coatings are being used increasingly for automobiles and aircraft. Urethane ester-type resins (also

¹Molecular weight value from number of molecules each multiplied by molecular weight and total divided by number of molecules

²Molecular weight value from sum of number of grams of material with a particular molecular weight each multiplied by its molecular weight and total divided by total number of grams

called urethane alkyds or uralkyds) are used primarily in architectural coatings. Two-component systems are used as high-performance coatings for maintenance and product finishes (Connolly *et al.*, 1986).

Silicone resins: Silicones are characterized by a siloxane backbone, e.g., $-\text{Si}-\text{O}-\text{Si}-\text{O}-$, with organic groups which determine the properties of the final polymer attached to the silicon atoms. The monomeric precursors of silicone polymers are mono-, di- and trisubstituted halosilanes (usually chlorosilanes). Monosubstituted silanols condense to highly cross-linked polymers, which are chiefly used in coatings. The degree of cross-linking and consequent physical properties are controlled by adjusting the ratio of mono- and disubstituted chlorosilanes. Alkyd resins with terminal hydroxyl groups can be condensed with silicones to produce hybrid polymers (Lowell, 1984).

Silicone resins are used to waterproof masonry and are blended with alkyds to formulate industrial maintenance coatings for storage tanks and other metal structures (Lowell, 1984).

Cellulose derivatives: Cellulose nitrate, commonly misnamed nitrocellulose, is the oldest cellulose derivative, first prepared in 1838 from cotton linters using a nitrating mixture of nitric and sulfuric acids (Jones, 1938; Sears, 1974). Before the early 1920s, only very high molecular-weight cellulose nitrate was available, which had limited utility in lacquers. The development of stable cellulose nitrate with lower viscosity after the First World War resulted in fast-drying lacquer coatings which were used extensively in automobile and furniture production. In the USA, three types of commercially available cellulose nitrates are distinguished by their nitrogen content and solubility. Each of these types is available in a variety of viscosity grades, which are a measure of the polymer chain length (Hamilton & Early, 1972; Brewer & Bogan, 1984).

Cellulose nitrate lacquers have also been formulated to contain resins, plasticizers, solvents and thinners. The resins that were employed initially with cellulose nitrate lacquers include shellac, sandarc, mastic and ester gums, which were added in amounts of about two-thirds of the weight of cellulose nitrate. Plasticizers are usually added at about 10% of the weight of cellulose nitrate. Camphor, which was first used in the USA in the mid-1800s, was replaced by castor oil; by the late 1920s, plasticizers such as triphenyl phosphate, tricresyl phosphate, dibutyl phthalate and butyl tartrate were being used increasingly. The principal solvents used initially with cellulose nitrate included ethyl, butyl and amyl acetates, acetone, 'diacetone alcohol', industrial spirit, ethanol and mixtures of alcohol with benzene or toluene and of alcohol with esters (Heaton, 1928; Hamilton & Early, 1972).

Another cellulose derivative, ethyl cellulose, is made by treating cellulose from wood pulp or cotton with a solution of sodium hydroxide to obtain primarily what is commonly referred to as 'alkali' or 'soda' cellulose. Further treatment with ethyl chloride under heat and pressure yields ethyl cellulose, which can be made in different viscosities. It is widely used in clear, dyed or pigmented lacquers for flexible substrates. Although less often used in paints, it has been formulated with silicone coatings to prevent pigment settling and sagging (Singer, 1957; Hamilton & Early, 1972).

Cellulose acetate is a linear high polymer that is obtained by first pretreating cellulose with a reduced amount of acetic acid to cause a certain amount of swelling, and then reacting it with acetic anhydride in the presence of sulfuric acid. Cellulose acetate lacquers are stable to light and heat and have good resistance to oils, greases and weak acids (Singer, 1957).

Methylcellulose, carboxymethyl cellulose and hydroxyethyl cellulose are water-soluble polymers that are used as thickeners in latex-based coatings. Cellulose acetate-butyrate is used as a resin modifier in automobile lacquers based on polymethylmethacrylate.

(c) *Solvents* (see also the monographs on solvents, pp. 43 *et seq.*)

Solvents are widely used to keep paints in liquid form so that they can be applied easily. Until the late nineteenth century, turpentine and alcohol were the only solvents of any importance. Since the early 1900s, the number of solvents has increased considerably to encompass initially a broad range of petroleum and coal-tar distillates, alcohols, esters, ketones, glycols and halogenated hydrocarbons and, more recently, synthesized glycol ethers and esters. A large variety of mixtures of these classes is also employed.

Solvents in the turpentine category are derived mainly from the resinous exudations of various species of pine and other conifers and consist essentially of mixtures of various terpenes such as α - and β -pinene (Heaton, 1928).

A petroleum distillate, known as 'white spirits' (see the monograph on some petroleum solvents, p. 43), which consists mainly of aliphatic, alicyclic and aromatic hydrocarbons, was introduced as a solvent in the paint industry in 1885. For many years it was regarded as a cheap adulterant for turpentine but, as its use developed, it attained recognition as a different solvent. In addition to white spirits, several other paint solvents are prepared from petroleum and coal-tars (see IARC, 1985b, 1987j). Coal-tar distillates were the original source of commercial quantities of solvents such as benzol (a mixture containing mainly benzene, with smaller amounts of toluene, light hydrocarbons and carbon disulfide), benzene (see IARC, 1982b, 1987k), toluene, xylene and solvent naphtha (Heaton, 1928).

(d) *Additives*

Additives are defined as those chemicals that perform a special function or impart a special property to paint. They are present at low concentrations, generally 0.2–10%, and include driers, thickeners, anti-skinning agents, plasticizers, biocides, surfactants and dispersing agents, antifoam agents and catalysts (Krivanek, 1982).

(i) *Surfactants*

Surfactants, which are classified into anionic, cationic, amphoteric and nonionic categories, are used as pigment dispersants in both nonaqueous and aqueous systems. Dispersants employed in nonaqueous systems include lecithin, zinc naphthenate, calcium naphthenate, copper oleate and oleic acid. Ionizable dispersants that are usually employed in aqueous coatings include tripotassium polyphosphate, tetrapotassium pyrophosphate, sodium salts of arylalkyl-sulfonic acids and sodium salts of carboxylic acids (Lowell, 1984).

In addition to pigment dispersion, surfactants are used in paints as emulsifying agents, protective colloids, wetting agents, thickeners and antifoaming agents. A number of water-

soluble resins and gums have been used as protective colloids or thickeners in emulsion paints. Water-soluble hydrophilic colloids include agents such as gum arabic, gum tragacanth, starch, sodium alginate, methyl cellulose, hydroxyethyl cellulose, polyvinyl alcohol, ammonium caseinate and sodium polyacrylate. The acrylate salts, casein and cellulose, have been widely used in acrylic paints, while the major thickeners for styrene-butadiene paints have been alkali-soluble proteins (soya bean proteins). Methyl cellulose and hydroxyethyl cellulose are common thickeners for polyvinyl acetate paints (Martens, 1964).

Noncellulosic thickeners used in latex paints include maleic anhydride copolymers, mineral fillers, such as colloidal attapulgite (see IARC, 1987l,m) and treated magnesium montmorillonite clays, natural products (e.g., alginic acid, casein and soya bean protein), polyacrylamides, polyacrylic acid salts and acid-containing cross-linked acrylic emulsion copolymers (Connolly *et al.*, 1986).

A recent partial list of surfactants employed in water-borne paints includes aluminium stearate, cellulose ethers, polydimethyl siloxanes, polyethylene, alkali metal phosphates and sodium dioctyl sulfosuccinate (Hansen *et al.*, 1987).

A variety of other surface-active agents are added to paints to control flow, levelling, sagging, settling and viscosity, including hydrogenated castor oils, lecithin, metallic soaps (e.g., linoleates, palmitates and stearates), treated montmorillonite clays, peptized oil gels, polyolesters, silicas and soap solutions (Connolly *et al.*, 1986).

(ii) *Driers*

Driers (siccatives) that have been used in water-borne paints containing unsaturated polymers (e.g., alkyds) to accelerate curing are principally metal salts (lead, calcium, cobalt, manganese, zirconium, barium, zinc and cerium-lanthanum) of naphthenic acid, tall oil acid, 2-ethylhexanoic acid and neodecanoic acid, generally at levels ranging from 0.3 to 0.8% (w/w; Hansen *et al.*, 1987). Cobalt-based driers are the most commonly used commercially and are active catalysts in both air drying and heat cure systems. Manganese is another major active drier. Other metal driers serve as auxiliary driers and are usually used in combination with cobalt and manganese. Lead (see IARC, 1980a, 1987n) driers were at one time the major auxiliary driers; however, legislation limiting the amount of lead that can be used in surface coatings has resulted in a sharply reduced use. The most suitable replacements for lead appear to be zirconium, calcium and cobalt-zirconium compounds (Connolly *et al.*, 1986). In addition, 1,10-phenanthroline has been employed at levels of 0.02% (w/w; Hansen *et al.*, 1987).

(iii) *Plasticizers*

The early use of plasticizers is illustrated by the incorporation of castor oil and glycerine into alcoholic spirit varnishes and of camphor into spirit varnishes and lacquers, as well as into cellulose ester enamels and lacquers in the late nineteenth and early twentieth centuries (Heaton, 1928). In 1912, triphenyl phosphate began to replace camphor for the plasticization of cellulose nitrate; later, tricresyl phosphate was used. The use of plasticizers was generally expanded by the mid 1920s with the introduction of di(2-ethylhexyl)phthalate (see IARC, 1982c) and dibutylphthalate in the mid-1930s (Sears, 1974).

Plasticizers that are generally added in quantities of up to about 2% include dibutyl-, diethyl-, diethylhexyl- and dioctylphthalates and, to a lesser extent, the low molecular-weight esters of adipic and sebacic acid, tributyl phosphate and castor oil. Polyester resins, including maleic residues, sulfonamides, triorthocresyl phosphate and chlorinated diphenyls, have been used occasionally (Piper, 1965; Krivanek, 1982).

(iv) *Biocides (fungicides, preservatives and 'mildewcides')*

Biocides are generally added to paint at low concentrations – less than 1% – for preservation in the tin. Each biocide formulation can contain several agents.

The function of a preservative is to retard the enzymatic degradation of cellulosic and other thickeners in latex paints in the tin. The function of a mildewcide is to retard the growth of fungi on applied exterior latex and solvent-based paints. These compounds are often the same, but are used in different quantities. Much less preservative is needed to preserve a latex paint than the amount of mildewcide required to retard mildew growth on an exterior paint (Connolly *et al.*, 1986).

Phenylmercury compounds (e.g., acetate, propionate, benzoate, dodecyl succinate and oleate) were previously used extensively as mildewcides. Although mercuric compounds are no longer employed in solvent-based paints in the USA because of legislative restrictions, they are still permitted in water-based paints. Biocides that have been employed include tributyltin oxide, chlorothalonil (see IARC, 1983a), 1,2-benzisothiazolin-3-one, carbendazim, benzyl alcohol mono(poly) hemiformal, 1-(3-chloroallyl)-tetra-aza-adamantane hydrochloride, 5-chloro-2-methyl-4-isothiazolin-3-one, dodecyl dimethylammonium chloride, 5,8,11,13,16,19-hexaoxatricosane, 3-iodopropynyl butyl carbamate, 2-methyl-4-isothiazoline-3-one, formaldehyde, sodium nitrite and sodium benzoate (Connolly *et al.*, 1986; Hansen *et al.*, 1987).

(v) *Antiskinning agents*

Antiskinning agents are added to paints to retard the formation of skin on the surface of the liquid coating, in either closed or open tins, without retarding the drying of the product. The principal antiskinning agents are oximes or phenol derivatives; the major oxime used is methyl ethyl ketoxime. Smaller quantities of butyraldoxime and cyclohexanone oxime are used. The phenol derivatives used are mainly methoxyphenol, *ortho*-aminophenol and polyhydroxyphenol. Minor quantities of cresols, guaiacol, hydroquinone (see IARC, 1977a), isobutoxysafrol and lignocol have also been used as antiskinning agents (Connolly *et al.*, 1986).

(vi) *Miscellaneous additives*

Other additives are employed in paints, including polymerization initiators such as benzoyl peroxide (see IARC, 1985c) and azobisbutyronitrile; antioxidants such as hydroquinones, phenols and oximes; ultraviolet light absorbers, luminescent and fluorescent materials and heat stabilizers (Connolly *et al.*, 1986).

1.2 History of the manufacture of paints and related products

(a) *History of paint manufacture*

The history of paints and other related products has been reviewed (Heaton, 1928; Jones, 1938; Singer, 1957; Martens, 1964).

Paint has existed from the earliest times – literally from the beginning of history. Water-based paint was used for pictorial and decorative purposes in caves in France and Spain as early as 30 000–15 000 BC. The earliest efforts of cavemen were expressed by daubing coloured mud on the walls of their caves. The addition of crushed berries, blood, milk, eggs, dandelion or milkweed sap and other natural materials, such as chalk, earth colours, charcoal and ashes, to early paints improved both adhesion and utility.

The earliest pigments were natural ores. By 6000 BC, calcined (fired) mixtures of inorganic components and organic pigments were employed in China. By 1500 BC, the Egyptians were using dyes such as indigo and madder to prepare blue and red pigments, and by 1000 BC they had developed a varnish from gum arabic that contributed to the permanence of their art. Although paint and varnish have existed since earliest times, it was not until the 1700s that commercial manufacture of paints began in Europe and the USA. The early manufacturers of paint ground their pigments on a stone table with a round stone.

Whitewash, which is essentially water-slaked lime, was used during the early history of many countries. Various materials, such as Portland cement, were added to improve the material and it gradually evolved into the present-day cement paints. Skimmed milk and, later, casein were added to improve adhesion and durability. The addition of pigments, whiting and clay and, finally, the replacement of lime traced the evolution from whitewash to casein paint. Drying oils were later added to casein paints to improve water resistance. Casein paints were continually improved and, by the 1930s, they contained highly refractive index pigments similar to those used in oil paints.

In the late 1800s, grinding and mixing machines were developed to enable manufacturers to produce large volumes of paint. However, it was not until the twentieth century, during the period from the mid-1920s to prior to the Second World War, when improvements in paint technology began to parallel more closely advances in chemistry, that paint became sophisticated with regard to the process of manufacture, its use and the methods of its application. The development of highly efficient paint-making equipment and an expanding scope of areas of utility, e.g., automobile, marine and architectural areas, contributed to these advances. Paint technology advanced most rapidly in countries where industrialization developed fastest, i.e., in western Europe and the USA, the greatest advances occurring after the Second World War (Brunner, 1978).

For centuries, it was common practice for painters to prepare their own products from pigments and oils (principally linseed oil). Little 'ready-mixed' paint was available until just before the beginning of the twentieth century, when a large number of factories were established, principally in Europe and the USA (Brunner, 1978).

Mixing was originally conducted in an apparatus known as a 'pug mill', the general construction of which was a cylinder fitted with a vertical shaft carrying arms. Until the introduction of machinery, grinding was generally accomplished by rubbing down the mixed paint

with a large 'muller' of glass or granite on a slab of similar material. Cone mills were later employed which were made in various sizes – small ones for hand use and larger sizes for factory use. Grinding machinery was introduced into paint manufacture in Europe around the late 1870s and developed enormously in design and efficiency during the twentieth century. Two- and, later, three-roller grinding mills were first introduced which greatly enhanced the speed and efficiency of grinding. Multiple-roll mills, e.g., four and five as well as combinations of edge-runner, horizontal mixers and roller mills, were all used in the 1920s for paint production and were employed until the advent of more modern plant methods. The conversion of paste paints to the liquid or 'ready-mixed' condition required a step called 'thinning' or the addition of a thinning vehicle. Vertical mixers were used for this purpose (Heaton, 1928).

(b) *History of lacquer manufacture*

The use of lacquer dates from about 500 BC when the Chinese and Japanese used the sap of the tree, *Rhus vernicefera*, to prepare lacquer for the ornamentation of woods and metals (Singer, 1957).

In the early manufacturing processes of the late eighteenth and early nineteenth centuries, the most frequent method of mixing the components of lacquers was by use of a slowly revolving churn that contained baffles. In older works, these were merely wooden barrels, but later they were specially designed and made of aluminium, since other metals are likely to affect the colour of clear lacquers due to the acidity of the solvents or resins used. At larger installations, a method that involved lifting up a container by means of a special lifting carriage to a battery of paddle stirrers was also used. After completion of dissolution, the container was covered and lifted by crane to its storage bench as standardized lacquers or stock solutions for further mixing (Heaton, 1928; Jones, 1938).

(c) *History of varnish manufacture*

The use of oil varnishes and resins was already well understood at the time of Theophilus the monk, who wrote about them in the eleventh century. The preparation of a drying oil by treating linseed oil with lime and litharge was described at an even earlier date by Eraclius in the ninth century (Barry, 1939). The travels of Marco Polo and the discovery of the New World brought a great variety of exotic gums and resins to Europe.

Towards the end of the eighteenth century, paints and varnishes were based on natural resins, e.g., copal and amber, and vegetable oils, e.g., linseed, walnut, hempseed and poppy seed oils. By the nineteenth century, fossil and semi-fossil gums replaced amber, which was by then scarce and expensive. These principal substitutes included gum arabic, gum elastic, copal, mastic and shellac (Singer, 1957). Early in the twentieth century, the first practical phenol-formaldehyde resins were developed. Research over the following two decades provided the basis for greatly improved varnishes.

Varnishes can be divided into three main types – oil, spirit or alcohol and water – of which the oil varnishes are the most important, such that the term 'varnish' generally implies oil varnish. The manufacture of oil varnishes involves the following operations: (i) 'gum running' or melting the resin; (ii) boiling the oil and mixing with the melted 'gum'; (iii) boiling the

varnish; (iv) introducing driers; (v) thinning the varnish; and (vi) maturing the varnish (Heaton, 1928).

1.3 Construction painting and paint products used

Paints that are used on architectural structures (indoor and outdoor surfaces) are comprised of primers or undercoats for walls and woodwork and mat, semigloss or gloss finishing coats. The primers and finishing coats differ primarily in pigment/vehicle balance and in additives and vehicle type. Primers (usually called 'primers/sealers') are used to seal the variable porosity of the substrate (e.g., wood) and to adhere to the substrate and to subsequent coats of paints. For most architectural uses, an alkyd-based primer/sealer is used (Schurr, 1981).

(a) *Exterior house paints*

Casein paints, which have been used since the mid-1800s, were continually improved so that, by the 1930s, they contained high-refractive index pigments. Later, a drying oil was added to casein to produce an emulsion paint (Martens, 1964). Traditionally, linseed oil and oleoresinous vehicles have accounted for the bulk of architectural (house) paints. Several other oils have been used, but to a much lesser degree than linseed oil, and often in conjunction with linseed oil. The more important have been tung oil, perilla oil, soya bean oil, fish oils, safflower oil and dehydrated castor oil. Modern oil-based house paints generally contain combinations of untreated drying oil (unbodied oil) and drying oil treated (polymerized) so that its viscosity is increased (bodied oil) (Schurr, 1974).

A wide variety of thinners and solvents has been employed traditionally – principally turpentine, white spirits, benzene and solvent naphtha. Turpentine was the accepted thinner until the 1930s, at which time white spirits were introduced for reasons of cost and odour (Schurr, 1974).

The classic house paint consisted of ~80% basic lead carbonate – white lead – and 11% raw or boiled linseed oil which contained a small amount of drier in the form of metallic soaps. The paint was thinned with additional oil for application. Gradually, other primer pigments and inert fillers were introduced into the basic lead-in-oil formula. These included zinc oxide, leaded zinc oxide, lithopone and, finally, titanium dioxide (see monograph, p. 307). The use of lead pigments has been increasingly curtailed because of legislation (Schurr, 1974, 1981).

In the late 1930s and 1940s, alkyd paints were gradually introduced, particularly in the dark colours, with a marked improvement in properties such as colour and gloss retention. In the 1960s, white alkyd house paints without lead pigments were marketed by a few paint companies; they usually contained phenylmercury oleate and other arylmercury derivatives as fungicides. By the 1970s, alkyd house paints were replacing oil house paints, the faster drying time of the alkyds being obtained with cobalt and calcium soaps.

Around 1957, the first exterior water-based house paints were introduced. Most of these were based on acrylic-type latexes, and the paint had excellent colour retention on exterior exposure. Since that time, water-soluble and emulsified linseed oil house paints

have been marketed which combine the advantage of an oil paint and a water-type paint in one product (Martens, 1964). Because of ease of application, cleaning ability with soap and water and good service, latex paints comprise most of the exterior paint market. Among the more common latexes are the acrylics, polyvinylacetate-dibutylmaleate copolymers, ethylene copolymers and acrylate copolymers (Schurr, 1974). Table 1 lists the ingredients of typical white house paints with oil and latex binders (Fisher, 1987).

Table 1. Examples of formulations of white house paints from the 1980s^a

Type of paint and ingredients	Weight (%)
<i>Oil-based</i>	
Titanium dioxide (anatase) } Titanium dioxide (rutile) }	12.7
Zinc oxide (acicular)	19.8
Water-ground mica	6.2
Magnesium silicate	23.2
Refined linseed oil	23.4
Bodied linseed oil	7.8
Lead and manganese soap solution	1.3
Mineral spirits	5.7
<i>Acrylic latex</i>	
Titanium dioxide (rutile)	20.2
Titanium dioxide (anatase)	0.8
Water-ground mica	2.5
Magnesium silicate	8.4
Calcium carbonate	6.9
Acrylic latex	41.9
Water	7.0
Cellulosic thickener	8.0
Nonionic emulsifier	0.9
Alkyl-aryl surfactant	0.2
Commercial defoamer	0.2
Ethylene glycol	2.1
Ammonium hydroxide	0.2
Organic mercurial fungicide	0.2
Pine oil	0.6

^aFrom Fisher (1987)

The main categories of organic pigments used in interior and exterior construction or architectural paints include phthalocyanine and monoazo dyes (Volk & Abriss, 1976).

(b) Interior paints

Until the early 1930s, the vehicles employed in interior paints were based on oils treated to increase viscosity (bodied oils) and heat-treated oils, usually combined with rosin,

ester gum or other natural gums. Most solvent-type paints for interior use contain some oil and dry by oxidation. Oils that have been used in interior compositions include bleached linseed oil, dehydrated castor oil, soya bean oil, tung oil and oiticica oil. The principal pigments used for interior white paints are titanium dioxide, zinc oxide and various carbonate and siliceous extenders which are used to control pigment volume and gloss. From about 1927, with the development of alkyd resins, a variety of architectural enamels for interior and exterior use was based on these resins. However, the bulk of enamels produced for interior use contained oil treated to increase viscosity (bodied oil) and/or varnish as the binder until after 1945 (Volk & Abriss, 1976).

Early water-based interior paints were alkyd-resin emulsions stabilized with large amounts of casein and other stabilizers. The alkyds used were generally long-oil vehicles and the paints generally had poor emulsion stability. Three types of latex polymers are used most commonly in the manufacture of latex paints: styrene-butadiene types, polyvinyl acetate types and acrylics. Copolymer blends of styrene and acrylate have also been employed, combining the most durable features of each monomer into a single polymer (Volk & Abriss, 1976).

After the Second World War, the excess capacity for manufacturing styrene-butadiene rubber (see IARC, 1979d) was adapted to make styrene-butadiene latexes that could be used in paint. These water-based latexes appeared in the USA around 1948 in interior wall finishes (Martens, 1964; Schurr, 1981). Although polyvinyl acetate latexes have been in existence since the late 1930s, they were used as adhesives rather than in paints until after the Second World War (Volk & Abriss, 1976).

The general categories of extender pigments that are used in latex paints include clays, calcium carbonates, silicates, diatomaceous earths, silicas, barytes and talcs (see IARC, 1987a,b,c,d). Along with latex, surfactants, pigments and several other additives are usually incorporated into the formulation to obtain a stable and satisfactory product, including thickeners, defoaming agents, freeze-thaw stabilizers, coalescents and pH adjusters. Although natural thickeners, such as casein, were formerly used, their use has decreased appreciably in recent years. The thickeners employed most commonly are cellulose – principally hydroxyethyl cellulose and methyl cellulose – polyacrylates, polyacrylamide, polyvinyl alcohols and many others. Ethylene and propylene glycols serve as freeze-thaw stabilizers. Coalescents are additives designed to optimize the coalescence of latex particles (Volk & Abriss, 1976) and include hexylene glycol, butyl cellosolve and butyl carbitol.

(c) *Masonry paints*

Casein-based paints applied to masonry and plaster surfaces were used in early construction work. These paints usually contained about 10% casein with some lime to insolubilize the casein after application (Martens, 1964). Latex-based primers/sealers are now often used for masonry surfaces. The latex vehicle is generally more resistant to alkali and permits evaporation of water from masonry surfaces without disruption of the film. Both alkyd and latex vehicles adequately seal porous surfaces (Schurr, 1981). Also, oil paints and styrene-butadiene copolymer, polyvinyl acetate emulsion, resin-emulsion and chlorinated rubber paints have all been used extensively on masonry surfaces.

Concrete floor coatings must possess good water resistance and adhesion over damp surfaces. Powdery concrete is first covered with a solvent primer. A satisfactory floor paint can be formulated using a styrene-butadiene latex fortified with an epoxy ester (Martens, 1964). Two examples of concrete floor enamels are presented in Table 2. Acrylic emulsion paints are widely used outdoors on concrete, stucco and cinder block because of their durability, adhesion and flexibility (Allyn, 1971).

Table 2. Examples of formulations of grey concrete floor enamels from the 1950s^a

Type of paint and ingredients	Weight (%) ^b
<i>Polystyrene-butadiene-based</i>	
Titanium dioxide (rutile)	17.6
Lampblack	0.5
Organic ester	0.2
Polystyrene-butadiene copolymer resin	27.0
Raw linseed oil	1.9
40% Chlorinated paraffins	4.7
High-flash naphtha	25.0
White spirits	24.4
<i>Chlorinated rubber-based</i>	
Titanium dioxide (rutile)	17.2
Lampblack	0.5
Organic ester	0.3
Chlorinated rubber	13.8
40% Chlorinated paraffins	3.2
Thermolysed tung oil	3.5
Alkyd resin	7.8
Soya lecithin	0.4
Dipentene	7.5
High-flash naphtha	14.1
Aromatic high-solvency petroleum solvent	24.6
White spirits	6.4
Antiskinning agent	0.1

^aFrom Singer (1957)

^bCalculated by the Working Group

Although cement paints are used on all types of masonry, they tend to be brittle and to 'powder off'. Cement paint typically contains white Portland cement, gypsum, calcium chloride and hydrated lime added to water. Fortified cements have been prepared using latexes such as styrene-butadiene, polyvinyl acetate and acrylic esters added to Portland cement in amounts ranging from 10 to 40% latex on a solid basis (Martens, 1964).

(d) Waterproofing paints

These paints are applied on the outside of unpainted concrete, brick, stucco, etc., and have been formulated in a variety of ways to include components such as wax, aluminium stearate and silicone resins. The earliest, simplest waterproofing formulations were of the wax type and consisted of paraffin wax, raw chinawood oil and white spirits. Stearate formulations consisted of aluminium stearate in white spirits, with occasional addition of paraffin wax. A significant advance in the manufacture of waterproofing paints in the mid-1950s involved the use of silicone resins. A typical formulation of silicone waterproofing contains silicone resin and xylene. Another commonly used product contains special silicones, such as sodium methyl siliconate, in aqueous solution (Singer, 1957).

1.4 Surface coating in the wood industry and products used

Shellac and other gums or resins, such as elemi, sandarac, manila and benzoin, dissolved in alcohol or spirits of wine were introduced by the French in the seventeenth century and have been used for a long time in Europe for finishing wood grain and for their quick-drying properties. By the beginning of the eighteenth century, use of varnishes on furniture increased rapidly, particularly in France and England, although the manufacture of varnishes was still incidental to the work of painters, decorators and gilders. By the mid- and late eighteenth century, varnishing of furniture was well established in Europe (Jones, 1938; Barry, 1939).

Four properties are considered to be essential in furniture varnish: quick, hard, tough drying (3–4 h); good sanding and polishing properties; good resistance to water, acids, alkalis, etc.; and good heat resistance (Singer, 1957). Table 3 gives the formulation of a best-grade furniture varnish used in the 1950s.

Table 3. Formulation of a furniture coating from the 1950s^a

Type of product and ingredients	Weight (%) ^b
<i>Phenolic resin-based furniture varnish</i>	
<i>para</i> -Phenyl phenol, pure phenolic resin	16.5
Modified phenolic resin, hard-oil type	11.0
China wood oil	17.3
Dehydrated castor oil	4.4
Xylene	16.5
White spirits	35.8
Cobalt naphthenate	0.3
Lead naphthenate	0.7
Antiskinning agent	0.3

^aFrom Singer (1957)^bCalculated by the Working Group

Four categories of wood stains were employed in the late nineteenth and early twentieth centuries: (i) water stains which consisted of water-soluble dyes or colouring agents

(e.g., potassium permanganate, potassium bichromate, pigments such as Venetian Brown, Bismark Brown); (ii) oil stains (e.g., brownish resins, bitumens, asphalts or pitches in white spirits, or petroleum tinted with oil-soluble dyes); (iii) naphtha stains, which consisted of naphtha-soluble dyes dissolved in coal-tar naphtha and containing a little resin such as coumarone or ester gum as a binder; (iv) and spirit varnish stains which consisted of methylated spirit-soluble dyes with a little alcohol-soluble gum and usually contained manila as a binder (Jones, 1938).

The range of organic dyes that are found in wood stains (see IARC, 1981) include rosaniline [magenta; see IARC, 1974b], nitrosine, indigo, amarantns, carmoisine (see IARC, 1975), croceine, rhodamine (see IARC, 1978a) and several CI solvent and acid dyes (Krivaneck, 1982).

Finishing operations for wood include staining, wash coating, filling (if necessary), sealing, sanding, application of one or two lustre coats and polishing. Two types of oil stains – soluble and suspended pigment type – impart the desired colour to wood. Wood stains are dissolved in a vehicle that enables the stain to soak into the wood rather than stick to its surface as a film. After the staining operation, a clear, thin coat of lacquer is often applied before application of a filler, which is called a 'wash coat'; it stiffens the protruding fine wood fibres and can be removed by light sanding. In some procedures, filler is used to fill the depressions before the sealer and finish coats are applied. These finishes are cured by solvent evaporation; finish coats usually contain cellulose nitrate (Lowell, 1984). Some typical sanding sealer formulations that have been used contain cellulose nitrate in ethanol, zinc stearate paste, maleic resin solution, castor oil, butyl and ethyl acetates, toluene and petroleum lacquer diluent (Singer, 1957).

Oil stains based on linseed oil and tung oil have been used in the finishing of furniture, since they protect against staining without leaving an apparent film on the surface. Varnishes based on urethane oils rather than oil resins are being used increasingly (Wicks, 1984).

Formulations of paint utilized for furniture are principally dependent on the end-use. Nursery furniture, for example, requires extremely hard, tough coatings containing non-toxic pigments. A wide variety of coatings has been used on furniture, including low-bake finishes based on urea-formaldehyde resins, polyurethane paints based on diphenylmethane diisocyanate and hexamethylene diisocyanate, and lacquers composed of ethyl cellulose or cellulose acetate butyrate combined with acrylic resins (Singer, 1957; Lowell, 1984).

1.5 Painting in the metal industry and paint products used

(a) *Metal primers, finish coats and corrosion inhibition paints*

Since iron and steel rust in time when in contact with moisture and oxygen, many products made with these metals are coated with rustproof primers and finish coats (Schurr, 1974).

Primers are vehicle-rich coatings intended for application as foundation and adhesion-promoting coats. Metal primers are used to form a firm adhesive bond with the surface and also serve as an impermeable barrier between the environment and metal surface. When

active rust prevention is essential, rust-inhibitive pigments that retard oxidation chemically are used.

Although there are many formulae for structural steel primers, red lead (Pb_3O_4 ; see IARC, 1980a) in a linseed oil vehicle has been used for a long time. Other formulations of red lead include combinations with alkyd resins and with red iron pigment. A typical red lead-iron oxide primer formulation is shown in Table 4. Zinc chromate (zinc yellow; a double salt of zinc and potassium and chromic acid) was introduced during the Second World War and is still used extensively. It is usually formulated as the basic pigment with an alkyd resin or linseed oil. Less zinc chromate is required to give the same protection as red lead, and zinc chromate is often combined with red iron oxide (Singer, 1957).

Because of restrictions on the use of lead and chromates, the pigments favoured in industrial maintenance coatings are now mainly zinc metal, zinc oxide, molybdates and phosphates (Schurr, 1981).

Finish coats cover the metal primer and seal it. Some metal products are covered by enamels which contain alkyd resins and dry by oxidation. The most durable coatings available are generally used on machinery and other industrial equipment and are based on epoxy or polyurethane resins which are cured by chemical reaction. Typical formulations are shown in Table 4.

(b) *Marine paints*

Paints for surfaces that are continuously immersed in seawater must be formulated with antifouling properties to resist the growth of marine fauna. Diverse species of hard and soft fauna which require a permanent anchorage in order to mature and reproduce form colonies on hulls.

The Phoenicians used copper on the hulls of their ships more than 3000 years ago. During the early nineteenth century, compounds (generally oxides) of copper, tin, lead, mercury and arsenic (see IARC, 1980c, 1987o) were the biocides used in antifouling paints, since these agents are effective against the broad range of organisms encountered in the marine environment. Biocides based on lead, mercury and arsenic are now prohibited from use in many countries (Brunner, 1978; Brady *et al.*, 1987).

Antifouling coatings based on derivatives of triphenyl or tributyl tin have been introduced during the past 15–20 years. In some coatings, an organotin compound, such as the acetate, chloride, fluoride or oxide, is simply mixed into the formulation. These coatings are known as 'free-association' coatings and are characterized by a leach rate of organotin which is quite high when the coating is new but rapidly diminishes until it is insufficient to prevent fouling. A more useful formulation is obtained when the organotin in 'copolymer' coatings is covalently bound to the resin of the coating and is released when the bond hydrolyses in seawater. Since organotin compounds do not prevent accumulation of algae on hulls, some commercial organotin coatings contain a small amount of cuprous oxide to control algae and grasses (Brady *et al.*, 1987).

Table 4. Examples of formulations of metal paints

Type of paint and ingredients	Weight (%)
<i>Red lead-iron oxide primer (from 1950s)^a</i>	
Stearated processed clay antisetting agent	0.3
Red lead (97%)	28.2
Red iron oxide (85% Fe ₂ O ₃ min)	7.0
Magnesium silicate	14.1
Mica, white, water-ground	3.5
Alkyd	27.9
White spirits	18.3
Cobalt naphthenate (6%)	0.1
Lead naphthenate (24%)	0.4
Antiskinning agent	0.1
<i>White epoxy powder paint (from 1980s)^b</i>	
Epoxy resin (1400 D)	60
Dicyanamide curing agent	4
2-Methylimidazole (accelerator)	1
Calcium carbonate (extender)	15
Titanium dioxide (pigment)	15
Acrylic polymer flow additive	1
<i>Water-based white epoxy enamel (from 1970s)^c</i>	
Epoxy resin emulsion (50% solids)	28.3
Polyamide resin curing agent (65% nonvolatiles)	20.0
Titanium dioxide	22.2
Hydroxyethyl cellulose	0.1
Water	29.4
<i>White polyurethane enamel (from 1980s)^d</i>	
Hydroxyl-functional resin (solids)	21.0
Dibutyl tin dilaurate (catalyst)	<0.1
Titanium dioxide (pigment)	19.0
Aromatic hydrocarbons	6.0
Propylene glycol monomethyl ether acetate	29.0
Polyisocyanate resin (solids)	8.0
Butyl acetate	1.0
Ethyl acetate	17.0

^aFrom Singer (1957); weight calculated by the Working Group

^bFrom Peltonen (1986); two-component product

^cFrom Allen (1972); two-component product

^dFrom Dupont (1988); two-component product

Most commercial antifouling paints contain a vinyl binder, although products with other binders are also available. Rosin or some other leaching agent is generally added to cuprous oxide formulations to permit its controlled release into seawater, where it is lethal to fouling larva forms. More recent developments in antifouling methods have involved use of a sheet material of black neoprene rubber impregnated with tributyl tin (Drisko, 1985). An antifouling coating used extensively by the the US Navy consists of cuprous oxide dispersed in a mixture of natural rosin and a vinyl chloride–vinyl acetate copolymer (Brady *et al.*, 1987).

More recent strategies have focused on nontoxic alternatives to antifouling paints. These include the use of fluoropolyurethane foulant–release coatings. One such formulation consisted of Desmodur–N–75 (an aliphatic polyisocyanate), polytetrafluoroethylene (38% by volume; see IARC, 1979c), titanium dioxide (see monograph, p. 307) and solvent (Brady *et al.*, 1987).

(c) *Automobile coatings*

The development of low–viscosity cellulose nitrate lacquers in the early 1920s revolutionized the painting of automobiles. Although these lacquers did not flow well and required an expensive buffing operation to obtain an acceptable gloss, their fast–drying characteristics permitted production line assembly and painting of automobiles for the first time (Lowell, 1984).

Cellulose nitrate lacquers were followed by the introduction of alkyd enamels to the automobile industry in the early 1930s. These compositions were usually modified with small amounts of amino resins to provide harder, more thoroughly cross–linked films. These were followed by the adoption of thermosetting acrylic enamels in which alkyds were replaced by acrylic copolymers containing hydroxyl groups which could still react with melamine modifiers (Lowell, 1984).

In the late 1950s, lacquers of greatly improved durability and gloss, based on polymethylmethacrylate or thermosetting acrylic enamels, were adopted by the automobile industry (Lowell, 1984).

Today, many new polymers, including maleic resins, amino resins (urea–formaldehyde and melamine–formaldehyde polymers), silicones, epoxides, polyesters and polyurethanes form the basis of highly diverse coating systems. In addition, nonaqueous dispersion lacquers and acrylic enamels have been developed. Steel used in automobiles is pretreated with a conversion coating (phosphating or bonderizing) to improve corrosion resistance and adhesion. In the traditional procedure, which is still employed to some extent, the solvent–borne primer was sprayed onto the automobile body shell followed by a surfacer which could be sanded. The primer and surfacer were often combined into a single adhesive formulation which could be sanded. The vehicles of primer–surfacers were combinations of oxidizing alkyd, epoxy and amino–formaldehyde resins, alkyd– and rosin–modified phenolic resins and others. Applications of the colour coats followed applications of the primer–surfacers combinations (Lowell, 1984).

Solvent–borne primers have been almost completely replaced by water–borne electro–deposited primers. The original anodic type has been largely replaced by the cathodic type which is superior in corrosion protection. The binders for cathodic deposition are typically

acid salts of amino-treated epoxy. The formulations contain polyepoxides or blocked polyisocyanates which cross-link the coating when it is baked. Prior to application of the top coat, a coat of solvent or water-borne epoxyester primer-surfacer is applied (Lowell, 1984).

Very solid top coats are being used increasingly, thus eliminating lacquer-type formulations. Versions of conventional thermosetting acrylic enamels that can be applied in about 40–50% volume solids are now available (Lowell, 1984).

A broad range of organic pigments is employed in automotive finishes. These include Hansa yellows (prepared from chloro- and nitroanilines and acetoanilides), diarylide yellow, nickel azo yellow (nickel (see IARC, 1976c, 1987p) chelate of diazotized 4-chloroaniline and 2,4-dihydroxy quinoline), lithol reds (precipitated azo pigments comprised of a family of the sodium, barium, calcium and strontium salts of the coupling product from diazotized 2-naphthylamine-1-sulfonic acid and 2-naphthol), yellow BON-maroon (manganese salt of the coupling product of diazotized 4-chloroanthranilic acid with 3-hydroxy-2-naphthoic acid), and naphthol reds and maroons (monoazo pigments such as the copper precipitation product from the coupling of diazotized 4-nitroanthranilic acid with Naphthanil RC). Other classes of nonazo organic pigments that have been employed in automotive finishes include quinacridones, thioindigos, perinones (diimides of naphthalene-1,4,5,8-tetracarboxylic acid), perylenes (diimides of perylene-3,4,9,10-tetracarboxylic acid) and anthraquinones (Fytelson, 1982).

Table 5 gives a typical formulation of a lacquer for automobile finishing.

Table 5. Formulation of an automobile paint from the 1970s^a

Type of paint and ingredients	Weight (%)
<i>Blue metallic lacquer</i>	
Acrylic resin	41.8
Methyl methacrylate (93%)	
Butyl acrylate (7%), comprising 40% nonvolatiles in toluene	
Cellulose acetate butyrate	4.4
Plasticizer	8.2
Aluminium pigment (65%)	1.8
Phthalocyanine blue pigment dispersion	1.4
Acetone	10.5
Methyl ethyl ketone	10.0
Xylene	4.0
Ethylene glycol monoethyl ether acetate	13.0
Mixed methyl esters of adipic and glutaric acid	4.9

^aFrom Williams (1977)

(d) *Aluminium paints*

Aluminium paints sold today in ready-mixed form are used as protective coatings on iron and steel, aluminium, magnesium and other metals, providing high resistance to moisture penetration and heat and a high reflectance for ultraviolet radiation. These paints con-

tain aluminium pigment in finely divided form and are formulated in two types – non-leafing and leafing. The former consists of aluminium ground to a powder that is dispersed in vehicles to give a metallic-grey finish. ‘Leafing’ is a phenomenon in which finely divided aluminium flakes rise to the surface and form a continuous metallic finish consisting of intermeshing aluminium flakes (Singer, 1957).

(e) *Coil coatings*

One of the most rapidly growing areas of industrial coating is coil coating. The coil stock consists of enormous rolls of thin-gauge steel or aluminium, which are coated at steel mills, aluminium mills or by specially equipped contractors. The coils are unwound, coated on high-speed roller coaters, heat cured and rewound. Binder compositions include alkyd-amino-formaldehyde combinations, vinyl chloride-vinyl acetate copolymers (see IARC, 1979a,b) and thermosetting acrylics, often modified with small amounts of epoxy, which produce coatings that are flexible, durable and adhesive. In the coil coating industry, solvent vapours are often collected and disposed of by incineration (Lowell, 1984).

1.6 Other painting trades and paint products

(a) *Traffic paints*

The major requirements for traffic (road) paints are fast and hard drying. The paints that are generally used contain a high pigment volume, fast-drying vehicles, such as resin combinations with low oil content or oil-free synthetic resins, and low-boiling solvents (e.g., petroleum fractions with distillation ranges of 100–150°C). Solutions of plasticized chlorinated rubber made of styrene-butadiene copolymers (containing an aromatic hydrocarbon solvent to maintain solubility) have been used. Glass spheres are added to formulations of reflecting paints (Lowell, 1984).

Conventional alkyd formulations (including both straight alkyd and alkyd/chlorinated rubber types) still account for most of the traffic paints used in the USA. However, there has been a significant increase in the use of more durable pavement marking materials, such as two-component polyester and epoxy systems and one-component hot extruded thermoplastic types (Connolly *et al.*, 1986).

Although traffic paints can be made in any colour, the most widely used are white (titanium oxide) and yellow (chrome yellow; Connolly *et al.*, 1986).

(b) *Fire-retardant paints*

Fire-retardant or intumescent paints, when applied to wood or other combustible surfaces, retard the spread of fire by foaming at elevated (but less than charring) temperatures. A number of intumescent formulae employed in the mid 1950s contained a chemical combination of polyol (e.g., pentaerythritol), a mono- or diammonium phosphate and an amide (e.g., dicyandiamide; Martens, 1964; Lowell, 1984). Certain pigments such as antimony oxide and borates are also added to enhance the fire-resistant properties of such paints. Other intumescent paint formulae have included polyvinyl acetate and acrylic latexes (Martens, 1964).

(c) *Aerosol colours*

A large variety of paints have been packaged in aerosol tins for touching up and painting small areas, hobby aircrafts and other such objects. The principal types of paint used are of alkyd composition, are thinned out to a low viscosity (generally with ketones and aromatic hydrocarbons) to allow atomization, and contain a gaseous propellant which is liquid under pressure (e.g., propane, butane and isobutane or dichloromethane, which has replaced dichlorofluoromethane in many countries). Other aerosol paint compositions include acrylic and cellulosic lacquers and epoxyester systems (Cannell, 1967; Lowell, 1984; Connolly *et al.*, 1986).

(d) *Paint and varnish strippers*

Before the advent of chemically-resistant synthetic paint vehicles, simple organic solvents, mixtures of solvents or solutions of caustic alkalis could be used to soften and strip most paints and various films (Downing, 1967).

Dichloromethane (see IARC, 1986c, 1987q) is the most widely used paint stripper base of the organic group. Other chlorinated hydrocarbons that have been used with dichloromethane are, in order of decreasing effectiveness, 1,2-dichloroethane (see IARC, 1979f), propylene dichloride, dichloroethyl ether and *ortho*-dichlorobenzene (see IARC, 1982d, 1987r). Other solvents that can soften paint films are, in (approximate) decreasing order of effectiveness, ketones (e.g., methyl ethyl ketone), esters, aromatic hydrocarbons (toluene (see monograph, p. 79) and naphthas), alcohols and aliphatic hydrocarbons (Downing, 1967).

Phenols and chloroacetic acids have been used in certain, specific situations, such as for stripping epoxy coatings. In addition, many less common solvents have been used, some primarily for specific applications, including 2-nitropropane (see IARC, 1982e), dimethylformamide (see monograph, p. 171), dimethyl sulfoxide, tetrahydrofuran and 1,1,2-trimethoxyethane (Downing, 1967).

The main inorganic compounds that have been used as paint strippers are alkalis, principally in the form of a boiling solution of sodium hydroxide and, to a lesser degree, potassium hydroxide and lime or soda ash (anhydrous sodium carbonate). Additives such as sequestering agents (e.g., gluconic acid and alkali metal gluconates), surfactants (e.g., sodium resinate, fatty acid soaps, sodium lignin sulfonate, alkylarenesulfonates and petroleum sulfonates), water-soluble activators (e.g., phenolic compounds and their sodium salts - cresol, chlorocresol, sodium pentachlorophenate) and solvents (e.g., monoethers of ethylene glycol and diethylene glycol) are often used to increase the stripping rates of inorganic paint removers. Paint removers that are used on steel, aluminium and other nonferrous alloys often contain corrosion inhibitors such as phosphates and chromates (Downing, 1967).

Molten and fused alkali baths are also employed to salvage ferrous metal parts with defective finishes. At temperatures up to 500°C, even heavy films of epoxy and silicone coatings can be removed rapidly (Downing, 1967).

2. Production and Use of Paint Products

2.1 Production

(a) Production processes

The modern manufacture of paints, which are generally made in batches, involves three major steps: (i) mixing and grinding of raw materials; (ii) tinting (shading) and thinning; and (iii) filling operations (US Environmental Protection Agency, 1979), as illustrated in Figure 1.

To produce a batch of paints, manufacturers first load an appropriate amount of pigment, resin and various liquid chemicals into a roller mill, which is a large, hollow, rotating steel cylinder. Mills for grinding primer or dark pigments are partly filled with steel balls that measure about 1–2 cm in diameter. Mills for grinding light colours usually contain flattened ceramic spheres (pebbles) that measure about 3–4 cm in diameter. Depending on the type of mill used, the grinding process lasts about 24 h or until the pigment has been ground to a sufficiently fine paste. After the pigment has been ground, more resin and solvent are added to the paste in the mill and the paste is 'let down', that is, pumped out of the mill through a strainer which removes the grinding media to a holding tank.

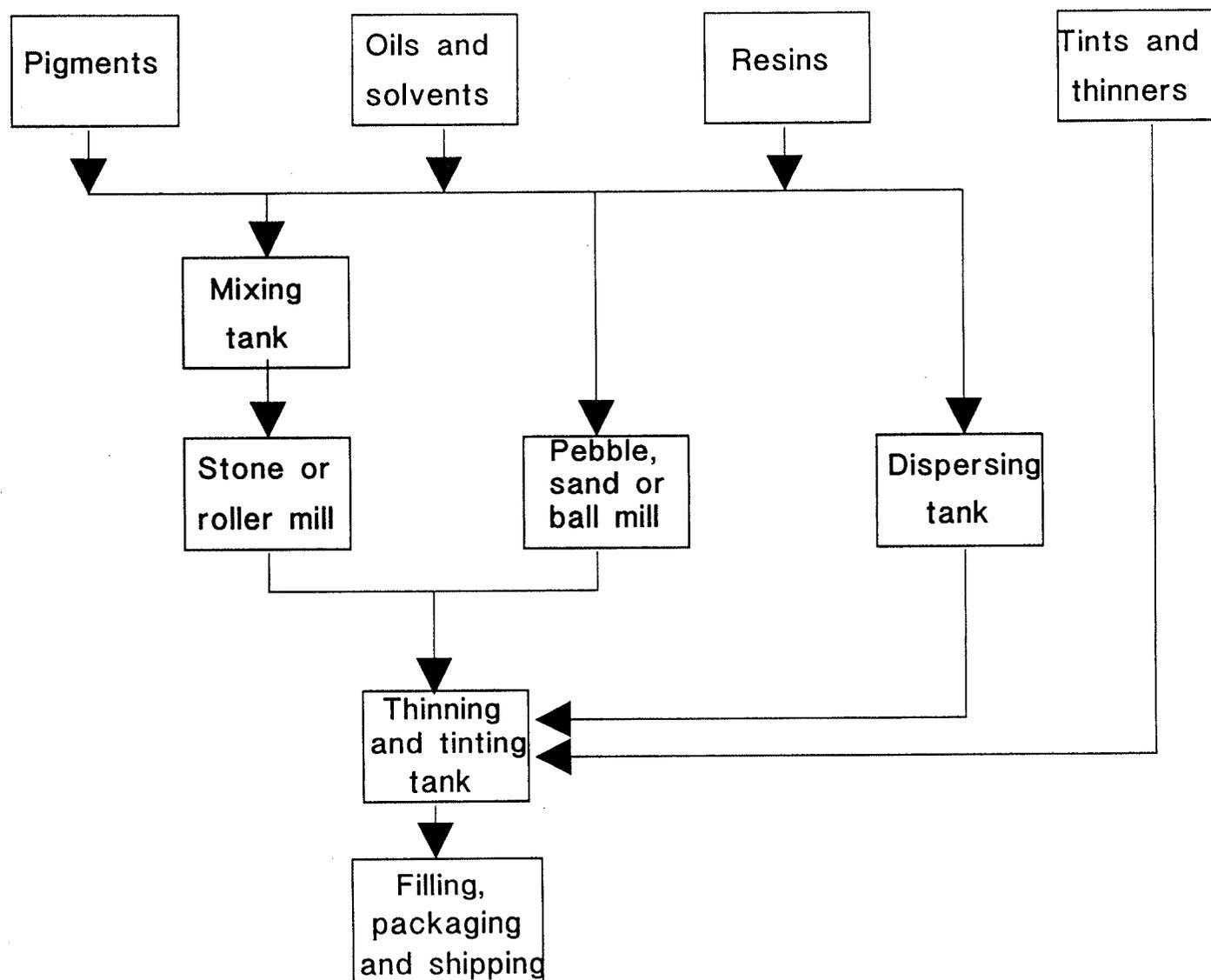
Until the 1930s, drying vegetable oils, primarily linseed oil, were used as binders in paints and as liquids for grinding. Since these oils were relatively poor pigment wetters, considerable energy was required for the grinding (dispersion) steps. Earlier dispersion techniques which involved pebble, steel ball or roller mills were replaced during the 1960s by high-speed equipment which was first used to dissolve large chips of pigment dispersed in solid binders. As pigment production and wetting characteristics improved, pigments were dispersed satisfactorily in high-speed dissolvers (Schurr, 1981).

The 'tinting' step involves comparing samples in the holding tank with colour standards. Small amounts of shading pastes, which are highly concentrated blends of ground pigments, and a vehicle are added as required to match the standard. After the batch has been shaded to specifications, it is thinned to the desired viscosity by the addition of solvent, filtered and poured into containers for shipment (Schurr, 1981).

The complexity of paint technology is indicated by the numerous types and number of raw materials required. A plant that produces a broad line of trade, maintenance and industrial paints requires over 500 different raw materials and purchased intermediates, including oils, pigments, extenders, resins, solvents, plasticizers, surfactants, metallic driers and other materials (Federation of Societies for Paint Technology, 1973).

The modern manufacture of unpigmented lacquers is generally a cold-cutting or simple mixing operation. For example, cellulose nitrate solutions are made by adding the nitrated cellulose from alcohol-wet cotton to the solvent mixture and agitating for 1–2 h in a paddle or turbine blade mixer. Alkyd resins, which are supplied in solution, can be added directly to the cotton-based solution. Hard resins may be dissolved separately, usually in toluene, and added as solutions, or the lumps may be dissolved directly in the cotton-based

Fig. 1. Process for manufacturing solvent-based paints^a



^aFrom US Environmental Protection Agency (1979)

solution by stirring. Liquid plasticizers are then simply poured or pumped into the mixing tank (Hamilton & Early, 1972).

In pigmented lacquer manufacture, the pigments are first dispersed in ball mills with plasticizers, such as dibutyl phthalate, after which natural or synthetic resins are added. Cellulose nitrate (or cellulose acetate) is then added and all the components are mixed in a vertical mixer or churn before the finished product is run off into containers (Browne, 1983).

Modern manufacture of varnish is carried out in jacketed and enclosed kettles or set pots, and the required high temperature is achieved by different methods, including the use of heat-transfer media (Browne, 1983).

(b) Production figures

Traditionally, two distinct types of coatings are produced – trade sale paints and industrial product finishes (Kline & Co., 1975).

Trade sale paints are sold through a variety of distribution channels to builders, contractors, industrial and commercial users and government units, as well as to the general public. These products are primarily for exterior and interior coatings for houses and buildings, although sizeable amounts of automobile and machinery refinishes, traffic paints and marine shelf-goods are also dispensed through trade sales outlets (Kline & Co., 1975).

Industrial product finishes or chemical coatings are produced to user specification and sold to other manufacturers for factory applications on such items as automobiles, aircraft, appliances, furniture and metal containers. They also include the category of industrial maintenance coatings, which are specially formulated and are used to maintain industrial plants and equipment (e.g., as resistance to corrosion). Within these major product lines, the paint industry produces thousands of different products for a broad spectrum of substrates, applications and customers (Kline & Co., 1975).

World production in 1971 of surface coatings by selected countries or regions is given in Table 6. North America was the largest producer and manufactured 4.5 million tonnes (31.6%), while western Europe produced 4.2 million tonnes (29.3%) and eastern Europe produced slightly over 3 million tonnes (21.6%; Kline & Co., 1975).

A more recent listing of paint production in the USA, Japan and western Europe is presented in Table 7. Japan is now the largest paint producer outside of the USA, followed by the Federal Republic of Germany, France, the UK and Italy. In 1986, US paint production was 967 million gallons [approximately 4340 thousand tonnes] (Reisch, 1987).

Estimated consumption of various resins, pigments and solvents in the USA in 1975, 1980 and 1985 is shown in Tables 8, 9 and 10. The major resins used in the production of paints are alkyd, acrylic and vinyl resins, which account for over 60% of total resin use in the USA. The main pigment was titanium dioxide and the major solvents aliphatic hydrocarbons, toluene and xylenes (see monographs, p. 125).

The number of paint manufacturers in the USA has declined steadily from about 1500 in 1963, to about 1300 in 1972 and 1000 in 1987 (Kline & Co., 1975; Layman, 1985; Reisch, 1987). In the UK, there have been similar reductions from about 500 paint manufacturers in the 1950s to only about 250–300 in 1985 (Layman, 1985).

Table 6. World production (in thousands of tonnes) of surface coatings by selected country or region in 1971^a

Region	Production	% Distribution
North America		
USA	4155	29.0
Canada	379	2.6
Total	4534	31.6
Europe		
Germany, Federal Republic of	1192	8.3
France	744	5.2
UK	614	4.3
Italy	420	2.9
Spain	207	1.4
Netherlands	192	1.3
Sweden	160	1.1
Belgium/Luxembourg	130	0.9
Austria	101	0.7
Denmark	84	0.6
Yugoslavia	80	0.5
Switzerland	68	0.5
Norway	66	0.5
Finland	46	0.3
Portugal	32	0.2
Turkey	27	0.2
Greece	20	0.1
Ireland	17	0.1
Total	4200	29.3
Eastern Europe (total)	3094	21.6
Asia		
Japan	1140	7.9
India	67	0.5
Israel	49	0.3
Hong Kong	26	0.2
Other ^b	304	2.1
Total	1586	11.1
Latin America^c	484	3.4
Oceania		
Australia	179	1.2
New Zealand	45	0.3
Other	4	<0.1
Total	228	1.6

Table 6 (contd)

Region	Production	% Distribution
Africa		
South Africa	81	0.6
Other	138	1.0
Total	219	1.5
TOTAL	14 345	100.0

^aFrom Kline & Co. (1975)

^bIncludes the Philippines and the Republic of Korea

^cIncludes Mexico

Table 7. Paint production (in thousands of tonnes) in selected countries in 1984^a

Country	Production
USA ^b	4432
Japan ^c	1803
Germany, Federal Republic of	1321
France	809
UK ^d	670
Italy	625
Spain	278
Netherlands	253
Sweden	189
Denmark	133
Belgium	131
Austria	126
Portugal	92
Finland	82
Switzerland	82
Norway	75

^aFrom Layman (1985), unless otherwise specified

^bFrom Connolly *et al.* (1986)

^cFrom Kikukawa (1986)

^dProduction in millions of litres

Table 8. Estimated consumption (in thousands of tonnes) of resins in paints and coatings in the USA^a

Resin	1975	1980	1985
Alkyd ^b	315	295	300
Acrylic	158	203	223
Vinyl	166	183	191
Epoxy ^c	38	70	87
Urethane	33	54	58
Amino	29	33	41
Cellulosic	24	27	24
Polyester ^b	11	33	62
Phenolic	11	11	12
Chlorinated rubber	6	8	8
Styrene-butadiene	11	7	6
Natural	9	8	7
Linseed oil	23	35	36
Other resins	77	61	66
Plasticizers	20	19	17
Total	931	1047	1138

^aFrom Connolly *et al.* (1986)

^bData for 1985 are not comparable to those for previous years, since it is probable that some of the resins reported as alkyds in previous years were actually polyesters.

^cIncludes unmodified epoxy resins plus hybrids (e.g., acrylate enhancement)

Table 9. Estimated consumption (in thousands of tonnes) of pigments in paints and coatings in the USA^a

Use and pigment	1975	1980	1985
Colours			
Titanium dioxide	323	354	393
Chrome	29	33	33
Iron oxide	43	53	57
Carbon black	8	8	9
Other coloured inorganic	7	5	5
Phthalocyanine	1	2	2
Other organic	7	7	8
Aluminium	10	11	11
Fillers			
Calcium carbonate	175	175	213
Talc	136	150	136
Clay	154	159	199

Table 9 (contd)

Use and pigment	1975	1980	1985
Fillers (contd)			
Silica	59	71	83
Barytes	34	33	34
Nepheline syenite and feldspar	19	34	35
Other extenders and fillers	17	20	26
Other			
Zinc oxide	10	12	12
Zinc dust	24	32	24
Lead (corrosion inhibiting)	11	8	5
Cuprous oxide	2	4	3
Other	3	6	6
Total	1072	1177	1294

^aFrom Connolly *et al.* (1986)

Table 10. Estimated consumption (in thousands of tonnes) of solvents in paints and coatings in the USA^a

Solvent	1975	1980	1985
Aliphatic hydrocarbons	533	456	433
Toluene	281	265	277
Xylenes	240	213	211
Other aromatic compounds	91	79	84
Butyl alcohols	50	59	68
Ethyl alcohol	82	84	95
Isopropyl alcohol	48	53	41
Other alcohols	25	26	29
Acetone	79	79	89
Methyl ethyl ketone	145	154	152
Methyl isobutyl ketone	47	48	50
Ethyl acetate	31	35	36
Butyl acetates	55	62	71
Propyl acetates	8	8	9
Other ketones and esters	61	68	75
Ethylene glycol	23	29	34
Propylene glycol	14	12	14
Glycol ethers and ether esters	109	120	136
Chlorinated solvents	6	10	21
Miscellaneous	16	16	15
Total	1944	1876	1940

^aFrom Connolly *et al.* (1986)

2.2 Use

(a) *Uses and application methods*

The various uses of paint products are described by type of resin in Table 11.

Paints are applied by direct contact or by deposition by atomization processes. The direct-contact category includes the familiar brushing and roller techniques, dipping, flow coating and electrodeposition. Deposition by atomization processes includes conventional spray, hot spray and electrostatic spray. Machine roller coating is used in the industrial application of paint to paper, plywood and metal sheets, as well as continuous coating of metal coils. Dip coating is used in large industrial operations (Browne, 1983; Lowell, 1984)

Probably the greatest advance made during the early 1900s in the field of paint technology was the introduction of the spray gun. Its advent helped in the introduction of cellulose nitrate lacquers and their application to automobile assembly line production. Electrostatic spraying was first introduced in the USA in the 1940s and later in the UK. Electrodeposition of paint, introduced during the 1960s, is an important milestone in industrial painting and has proven especially advantageous for painting automobile bodies and other parts because of its superior corrosion resistance. In this technique, the coating is an aqueous dispersion of low solid content. The binder particles carry ionized functional groups which may be positive or negative, thus having either anodic or cathodic deposition. The anodic type typically uses amino- or alkali-solubilized polycarboxylic resins and the cathodic type, salts of amine-treated resins, such as epoxy resins (Brewer, 1984; Lowell, 1984).

(b) *Use patterns*

Use of paints in the major markets in the USA in 1985 is presented in Table 12. Distribution of use of resins and the other main components of paints in the USA in 1985 is shown in Table 13.

2.3 Exposures in the workplace

(a) *Introduction*

No data on the numbers of paint production workers or painters worldwide were available to the Working Group. According to a 5% census sample of the US population in 1970 (National Institute for Occupational Safety and Health, 1970), paint manufacturers employed approximately 62 000 workers. Extrapolating from the proportion of world production of surface coatings contributed by the USA (see Table 6), it can be estimated that the total number of paint production workers in the world is approximately 200 000. According to the same US census survey, there were 362 000 construction/maintenance painters and 106 000 painters/paperhangers/decorators in the USA. On the basis of these figures, it can be similarly estimated that the number of painters worldwide must be at least several million.

Table 11. Uses of polymer systems in industrial coatings^a

Polymer systems	Coil	Metal	Appli- ance	Furni- ture	Hard- board	Lumber and plywood	Marine	Mainte- nance	Auto- mobile manu- facture	Auto- mobile refinish	Tins	Paper- board
Natural and modified polymers												
Drying oils				+	+	+	+	+			+	
Cellulose esters		+		+		+			+	+		+
Cellulose ethers				+						+		
Condensation systems												
Alkyd resins	+	+	+	+	+	+	+	+	+	+	+	+
Polyesters, high molecular weight	+	+	+		+	+						
Amino resins	+	+	+	+	+	+			+	+		
Phenolic resins	+	+	+				+	+	+	+	+	
Polyamides		+					+	+			+	+
Polyurethanes				+	+		+	+	+		+	+
Epoxy resins	+	+	+	+	+		+	+	+		+	
Silicones		+	+				+	+			+	
Vinyl polymers and copolymers based on:												
Butadiene								+			+	
Acrylic or methacrylic ester	+	+	+	+	+		+	+	+	+	+	+
Vinyl acetate				+	+	+	+	+				
Vinyl chloride	+	+	+	+	+	+	+	+	+		+	
Vinylidene chloride							+	+				+
Styrene		+	+		+			+	+	+	+	+
Vinyl acetal or butyral	+	+		+				+			+	
Fluorocarbons	+											
Resin combinations												
Acrylic and amino	+	+	+	+	+				+		+	+
Acrylic and epoxy		+	+						+		+	
Acrylic and silicone	+	+						+				
Alkyd and amino	+	+	+	+	+				+	+	+	+
Alkyd and acrylic		+	+	+				+	+	+	+	
Alkyd and epoxy		+	+					+	+			
Alkyd and silicone	+	+	+									
Polyester and epoxy		+	+					+	+		+	+
Polyester and silicone	+	+			+							
Cellulose ester and urethane				+								
Alkyd, acrylic and amino					+							
Polyester and amino											+	+
Phenolic and epoxy							+	+			+	
Epoxy and amino											+	
Phenolic and amino											+	
Alkyd and vinyl chloride polymers							+	+				

^aFrom Lowell (1984)

Table 12. Consumption of paints and coatings by major market in the USA, 1985^a

Paints and coatings	%	% of total
Architectural coatings		46
Water-based	73	
Solvent-based	27	
Product finishes		38
Miscellaneous ^b	32	
Containers	15	
Wooden furniture	14	
Automotive	12	
Machinery	10	
Metal furniture	7	
Coils	5	
Wood flat stock	4	
Special purpose coatings		16
Traffic	28	
Auto refinishes	25	
Special maintenance	19	
Aerosols	10	
Other ^c	18	

^aFrom Connolly *et al.* (1986)

^bIncludes appliances, other transportation, marine, paper and foil, and other

^cIncludes paints for roofs, bridges, marine shelf goods, metals and others

A wide range of potential occupational health hazards is present in relation to the manufacture and use of paints, varnishes and lacquers. Coatings are complex mixtures containing a variety of groups of substances, such as organic solvents, organic and inorganic pigments, extenders, resins and additives such as catalysts, surfactants, driers, plasticizers and biocides. Each of these categories covers a range of tens or hundreds of individual chemical compounds (Connolly *et al.*, 1986). It has been reported that over 3000 individual paint components are used worldwide.

Occupational exposure results predominantly from the inhalation of gases and vapours, mainly organic, from solvents, binders and additives, of mainly inorganic pigment dusts and of complex inorganic and organic mixtures such as dusts from dried coatings and mists generated during the spraying of paint. The other major route of occupational exposure is through cutaneous contact with the various paint compounds, many of which can be absorbed through the skin. Ingestion related to personal work habits constitutes another potential route of entry.

Table 13. Distribution of use of components of paints in the USA in 1985^a

Use	Proportion by use (wt% of total)					Total (wt% of grand total)
	Resins	Pigments	Additives	Solvents	Water	
Product finishes						
Wood furniture and fixtures	25.7	2.9	0.1	66	5.3	4.4
Wood mat stock	38	20	0.6	24	17	1.7
Metal furniture and fixtures	32	25	0.1	36	6.7	2.8
Containers and closures	38	6.2	0.3	43	13	4.8
Sheet, strip and coil	32	26	0.1	33	8.1	2.3
Major appliances	37	24	0.1	33	5.4	1.4
Other appliances	39	22	0.1	35	3.5	0.8
Automobile						
Topcoat	32	16	0.05	46	5.7	1.7
Primer	23	24	0.3	29	23	1.7
Underbody components and parts	26	18	0.2	35	21	1.1
Trucks and buses	31	16	0.1	27	25	1.2
Railroad	25	25	0.1	33	17	0.4
Other transportation	38	25	neg	38	neg	0.4
Machinery and equipment	30	27	0.2	38	5	4.2
Electrical insulation	51	0.0	0.0	49	neg	1.1
Paper, film and foil	39	2	0.1	43	17	1.0
Other product finishes	27	17	0.2	40	16	5.2
Total	31	17	0.2	41	11	36.8
Architectural coatings						
Interior water-based						
Mat	14	40	2	1.6	43	16.5
Semigloss and gloss	18	23	2	9.5	48	4.5
Other	18	21	1.5	0.2	57	1.3
Interior solvent-based						
Mat	21	58	0.2	30	0.0	1.1
Semigloss and gloss	27	41	0.1	32	0.0	2.6
Varnish	33	1.4	0.2	27	0.0	0.7
Other	26	21	0.2	43	0.0	0.7
Exterior water-based						
Mat (house paints)	19	35	25	4.2	39	11.2
Trim	20	23	2.6	3.3	51	0.9
Stains	17	19	1.5	3.5	59	1.4
Other	17	24	1.7	5	52	0.8
Exterior solvent-based						
Mat (house paints)	28	38	0.5	33	0.0	2.1
Enamel	42	26	0.3	32	0.0	2.4
Primer	25	37	0.7	37	0.0	0.7
Varnish	58	0.0	0.4	42	0.0	0.3
Stains	41	0.0	0.4	59	0.0	1.2
Other	24	40	0.5	36	0.0	0.3
Total	20	33	1.7	12	33	48.5

Table 13 (contd)

Use	Proportion by use (wt% of total)					Total (wt% of grand total)
	Resins	Pigments	Additives	Solvents	Water	
Special-purpose coatings						
Maintenance	36	23	0.2	33	8.2	2.8
Marine						
Pleasure	50	0.0	0.5	50	0.0	0.04
Commercial and maintenance	35	29	0.1	35	0.5	1.1
Automobile refinishing	23	22	0.2	55	neg	3.2
Traffic paints	15	59	0.1	26	0.9	5.6
Aerosol	15	5.5	0.1	80	neg	1.1
Other	21	46	0.2	32	0.1	3.2
Total	23	38	0.1	37	1.8	16.4
Grand total (thousands of tonnes)	1138	1294	42	1217	884	4536

^aFrom Connolly *et al.* (1986); neg, negligible

Workers in the painting trades may also be exposed to a number of chemical agents originating from other operations that they or fellow workers are involved in, such as cleaning and preparing by chemical or mechanical means the object to be painted or cleaning themselves and the painting equipment.

The main substances to which workers may be exposed are listed in Table 14. The main occupational agents for which quantitative exposure data are available are presented in the following sections, covering the major paint trades.

Exposure to solvent mixtures is often described in the following sections using a summary measure, the cumulative exposure index (CEI), i.e., the sum of ratios of various measured levels to the respective occupational exposure limits. If this index exceeds unity, the combined exposure to different components of a solvent mixture is considered to exceed the recommended exposure limit. The values of the CEI are not always comparable because the exposure limits may vary with country and time.

In some painting operations, personal protective equipment is worn. However, it is common industrial hygiene practice to determine potential exposure by monitoring the breathing zone outside such protective gear. The results reported are thus not necessarily actual personal exposures.

Table 14. Main substances (and classes of substances) to which workers may be exposed in the painting trades^a

Material	Principal uses or sources of emissions	IARC Monographs ^b
Acrylates (e.g., ethyl acrylate, methyl methacrylate)	Acrylic resins, ultraviolet curing paints	IARC (1979c)
Acrylic resins	Binders	IARC (1979c)
Alcohols, aliphatic (e.g., methanol, isopropanol, <i>n</i> -butanol)	Solvents (lacquers), paint removers	
Alkalis (e.g. sodium hydroxide, potassium hydroxide)	Paint removers	
Alkyd resins	Binders	
Aluminium, powder	Pigment	
Amides, aliphatic (e.g., dimethylformamide)	Solvents	This volume
Amines (mono), aliphatic (e.g., diethylamine) and alkanolamines (e.g., 2-amino-2-methyl-1-propanol)	Water-based paints	
Amines (poly), aliphatic (e.g., diethylenetriamine)	Curing agents (epoxy resins)	
Amines, aromatic (e.g., <i>meta</i> -phenylenediamine, 4,4-methylenedianiline)	Curing agents (epoxy resins)	IARC (1978b) IARC (1986d)
Amino resins (e.g., urea-formaldehyde resins, melamine-formaldehyde resins)	Binders	IARC (1982a)
Ammonia	Water-based paints	
Anhydrides, organic (e.g., maleic anhydride, phthalic anhydride, trimellitic anhydride)	Alkyd resin synthesis, curing agents (epoxy resins)	
Antimony compounds (e.g., antimony trioxide)	Pigments, fire retardant pigments	This volume
Arsenic compounds (e.g., copper aceto-arsenate)	Antifouling agents	IARC (1980c)
Asbestos	Filler, spackling and taping compounds, talc	IARC (1977b)
Barium compounds (e.g., barium sulfate, barium carbonate)	Pigments	
Benzoyl peroxide	Catalyst	IARC (1985c)
Bisphenol A	Epoxy resins	
Cadmium compounds (e.g., cadmium sulfide, cadmium sulfoselenide)	Pigments	IARC (1976c)
Calcium compounds (e.g., calcium sulfate, calcium carbonate)	Fillers	
Camphor	Plasticizer	
Carbon black	Pigment	IARC (1984)
Cellulose ester resins (e.g., cellulose nitrate, cellulose acetate)	Binders	
Chloracetamide	Fungicide (water-based paints)	
Chromium compounds (e.g., chromic oxide, chromates)	Pigments	IARC (1980a)
Chlorofluorocarbons	Spray-can paint propellants	IARC (1986e)
Clays (e.g., bentonite)	Fillers	

Table 14 (contd)

Material	Principal uses or sources of emissions	IARC <i>Monographs</i> ^b
Coal-tar and asphalt	Special waterproof coatings (ships, tanks, pipes)	IARC (1985b)
Cobalt compounds	Pigments, driers	
Copper and copper compounds (e.g., bronze powder, cuprous oxide)	Pigments, antifouling agents	
Dyes and pigments, organic (e.g., aromatic azo dyes, phthalocyanines, rhodamine)	Pigments	IARC (1974b, 1978a)
Epichlorohydrin	Epoxy resins	IARC (1976b)
Epoxy resins	Binders	IARC (1976a)
Esters, aliphatic (e.g., ethyl acetate, isopropyl acetate)	Solvents	
Ethers, aliphatic (e.g., isopropyl ether, tetrahydrofuran) and glycol ethers (e.g., methylcellosolve)	Solvents	
Formaldehyde	Amino resin varnishes, biocide (water-based paints)	IARC (1982a)
Gasoline	Solvent	IARC (1989a)
Glycidyl ethers (e.g., <i>n</i> -butyl glycidyl ether and bisphenol A diglycidyl ether)	Epoxy resin diluents and constituents	This volume
Glycols (e.g., ethylene glycol)	Polyester resins, water-based paints	
Hydrocarbons, aliphatic (e.g., hexanes, heptanes)	Solvents (naphthas, white spirits)	
Hydrocarbons, aromatic (e.g., benzene, toluene, xylenes, trimethylbenzene)	Solvents (naphthas, white spirits), paint removers	IARC (1982b); this volume
Hydrocarbons, chlorinated (e.g., dichloromethane, 1,1,1-trichloroethane, carbon tetrachloride, trichloroethylene)	Solvents, paint removers, metal degreasers	IARC (1979g,h,i; 1986c)
Hydrochloric acid (hydrogen chloride)	Catalyst (amino resins)	
Iron compounds (e.g., iron oxides, ferric ferrocyanide)	Pigments	IARC (1972)
Isocyanates (e.g., 1,6-hexamethylene diisocyanate, toluene diisocyanate)	Two-component polyurethane resins	IARC (1986b)
Isothiazolones (e.g., 1,2-benzisothiazolin-3-one)	Biocides in tinned foods	
Kerosene	Solvent	IARC (1989b)
Ketones, aliphatic (e.g., acetone, methyl ethyl ketone, cyclohexanone, isophorone, diacetone alcohol)	Solvents, lacquers, paint removers	
Lead compounds (e.g., lead chromate, lead oxides, basic lead carbonate, lead naphthenate)	Primers, pigments, driers	IARC (1980a)
Magnesium compounds (e.g., magnesium carbonate)	Fillers	
Manganese naphthenate	Drier	
Mercury compounds (e.g., mercuric oxide, phenyl mercuric acetate)	Fungicides (water-based paints)	
Methyl cellulose	Thickener (water-based paints)	

Table 14 (contd)

Material	Principal uses or sources of emissions	IARC Monographs ^b
Mica	Filler	
Molybdenum compounds (e.g., lead molybdate)	Pigments	
Nickel, metal powder	Pigment	IARC (1976c)
Nitroparaffins (e.g., nitroethane, 2-nitropropane)	Solvents	IARC (1982e)
Oils, vegetable (e.g., linseed oil, tung oil)	Binders	
Oximes (e.g., methyl ethyl ketoxime)	Anti-oxidants, anti-skinning agents	
Petroleum solvents (e.g., Stoddard solvent, VM & P naphtha)	Solvents, paint removers	This volume
Phenol	Phenol-formaldehyde resins, paint remover (formerly)	This volume
Phenol-formaldehyde resins	Binders	
Phenols, chlorinated (e.g., pentachlorophenol)	Fungicides (water-based paints)	IARC (1979j)
Phosphates, organic (e.g., tricresyl- <i>ortho</i> -phosphate, tributyl phosphate)	Plasticizers	
Phthalate esters (e.g., dibutyl phthalate, dioctyl phthalate)	Plasticizers	IARC (1982c)
Polychlorinated biphenyls	Plasticizers	IARC (1978c)
Polycyclic aromatic hydrocarbons	Special waterproof coatings (ships, tanks, pipes)	IARC (1983b)
Polyester resins	Binders	
Polyurethane resins	Binders	IARC (1979k)
Polyvinylacetate resins	Binders	IARC (1979b)
Pyrolysis fumes	Removal of paint by burning; heat-curing operations	
Rosin	Binder	
Rubber, synthetic (e.g., butyl rubber, styrene-butadiene rubber)	Binders (special paints, water-based paints)	IARC (1982f)
Shellac resin	Binder	
Silica, amorphous (e.g., diatomaceous earth)	Filler	IARC (1987b)
Silica, crystalline (e.g., quartz)	Filler, sand-blasting operation	IARC (1987b)
Silicates (e.g., sodium silicate, aluminium silicate)	Fillers	
Stearates (e.g., aluminium, zinc stearates)	Soaps, flattening agents	
Strontium compounds (e.g., strontium chromate, strontium sulfide)	Pigments	IARC (1980b)
Styrene	Polyester resins	IARC (1979d)
Styrene oxide	Diluent (epoxy resins)	IARC (1985d)
Sulfuric acid	Metal cleaner	
Talc	Filler	IARC (1987c)
Tin, metal powder	Lacquers (tinplate containers)	
Tin, organic compounds (e.g., tri- <i>n</i> -butyltin oxide, dibutyltin laurate)	Antifouling agents, catalysts	
Titanium dioxide	Pigment	This volume
<i>para</i> -Toluenesulfonic acid	Catalyst (amino resins)	
Turpentine	Solvent	

Table 14 (contd)

Material	Principal uses or sources of emissions	IARC <i>Monographs</i> ^b
Vinyl acetate	Polyvinylacetate resins	IARC (1986a)
Zinc and compounds (e.g., zinc metal powder, zinc oxide, zinc chromate)	Pigments, catalysts, bodying agents	IARC (1980b)

^aFrom Sterner (1941); Piper (1965); Phillips (1976); O'Brien & Hurley (1981); O'Neill (1981); Dufva (1982); Krivanek (1982); Ringen (1982); Adams (1983); Selikoff (1983); National Institute for Occupational Safety and Health (1984); Swedish Work Environment Fund (1987) and previous sections

^bSee also *IARC Monographs Supplement 7*

(b) *Manufacture of paints and related products*

The manufacture of paints and related products such as varnishes, lacquers, enamels and paint removers involves the handling and processing of a complex array of raw materials, e.g., pigments, extenders, solvents, binders and additives, described in section 1, implying overall potential worker exposure to hundreds of chemicals (National Institute for Occupational Safety and Health, 1984). Furthermore, raw materials are often subjected to chemical changes such as during polymerization and cooking, thus creating a variety of new hazards.

The potential for occupational exposure depends largely on the basic types of products being manufactured, the degree of automation of the manufacturing process, the availability of exposure control measures and the nature of the specific job held. Various job classification systems have been developed for the paint manufacturing industry. Workers have thus been regrouped according to the basic product made – water-based paints, solvent-based paints, lacquer and vehicle – and to functions – pre-batch assembler, mixer, tinter, filler, tank and tub cleaner, reactor operator, varnish cooker, filter press operator (Morgan *et al.*, 1981). Additional functions are raw materials handler, laboratory personnel and others such as packagers, maintenance personnel, shippers and warehouse workers (National Institute for Occupational Safety and Health, 1984).

Heavy exposures, both by inhalation and skin contact, occur specifically in operations that can involve manual handling procedures such as weighing dry ingredients (pigments, extenders, resins, additives), loading them into mixing equipment, adding solvents to mills, and cleaning equipment (mixers, mills, reactors, kettles, tanks, filters). Additional exposure to solvents occurs in thinning, tinting and shading procedures, filling operations and filtering of varnishes. The cooking of varnishes may produce emissions of various aldehydes such as acrolein, of phenol, ketones, glycerine and fatty acids as well as dusts or vapours of maleic, phthalic and fumaric anhydrides during the loading of kettles. The production of powder coatings can be associated with significant exposure to dust from resin powders, pigments, curing agents and other additives. In the manufacture of radiation-curable coatings, exposures may occur to monomers such as ethyl acrylate, other acrylates and photoinitiators. Caustic solutions may be used in the cleaning of dispersion equipment (National Institute for Occupational Safety and Health, 1984). In general, important opportunities for exposure

result from the presence of spills and the continuous spattering from machines (Adams, 1983).

(i) *Exposure to solvents*

Because of their volatility, solvents are ubiquitous air contaminants in paint manufacturing industries.

Exposure levels measured for various categories of workers in nine Swedish companies and reported as the sums of standardized concentrations are summarized in Table 15. High concentrations of solvents were found in all operations, the worst situation being manual cleaning of equipment with solvents. Local exhaust ventilation was common, and respirators were not often used. Of the 14 types of solvents monitored, the most common were xylene, toluene, butanol and esters (Ulfvarson, 1977).

Exposure to organic solvents was measured in the breathing zone of 17 Swedish male paint industry workers presumed to have the highest exposure of 47 workers employed in seven factories in Sweden, by collecting air with battery-driven syringes and analysing with two portable gas chromatographs. The median exposure values were (mg/m^3): xylene, 111 (16 persons); toluene, 11 (16 persons); isobutanol, 5 (15 persons); ethylacetate, 20 (14 persons); *n*-butylacetate, 14 (13 persons); ethanol, 13 (13 persons); *n*-butanol, 7 (13 persons); methylacetate, 12 (eight persons); dichloromethane, 719 (three persons); white spirits, 45 (three persons); and isopropanol, 129 (one person) (Haglund *et al.*, 1980).

Overall solvent exposure of workers known to be exposed to toluene was measured in seven paint manufacturing companies in New Zealand. Mean total levels of solvents ranged from 19 ppm in one company (five workers sampled) to 130 ppm in another one (three workers sampled), with individual values ranging from 7 to 297 ppm. Toluene, xylene and ethylbenzene were found in the atmosphere of all seven plants; the frequencies of other solvents were as follows: heptane, 6/7; *n*-hexane and methylethylketone, 5/7; acetone, 4/7; pentane, methylisobutylketone, ethanol and *n*-butylacetate, 3/7; and isopropanol, 1/7 (Winchester & Madjar, 1986).

In another study in Sweden, 47 employees of seven paint manufacturing industries, known to be exposed to solvents and including nine manual cleaners of paint mixing equipment, were surveyed for exposure to 12 solvents. The results are summarized in Table 16. The main exposures with regard to both frequency and weight were to xylene and toluene. Outstandingly high exposures occurred during the manual cleaning of equipment (Lundberg & Håkansson, 1985).

In a study on the effects of long-term exposure to solvents in the paint industry in Sweden, overall solvent exposure in a large paint manufacturing company was estimated for various work tasks over three historical periods. The results are presented in Table 17 in terms of the CEI, i.e., the sum of the ratios of the various exposure levels to the respective Swedish occupational standards in 1982 for the various solvents (Ørbaek *et al.*, 1985).

Table 15. Exposure levels (personal breathing-zone samples) to combined organic solvents during various paint manufacturing operations^a

Operation	Sampling condition		Combined exposure ^b		Main solvents ^c	No. of samples in which solvent found
	No. of samples	Sampling time (min)	Mean	Range		
Charging solvents	33	4-43	2.0	0.2-16	Xylene	16
					Mesitylene	4
					Toluene	4
					Styrene	2
					Butanol	9
Pigment dispersion	18	9-66	1.5	0.2-4.4	Xylene	13
					Butanol	4
Tinting, thinning	14	15-32	0.9	0.1-2.0	Xylene	11
					Butanol	3
Can filling, paints	39	11-32	1.3	0.02-6.6	Xylene	23
					Alkanes	4
					Butanol	7
					Benzene	4
					Toluene	6
Can filling, thinners	14	9-20	1.8	0.1-7.4	Toluene	3
					Xylene	5
					Trichloroethylene	3
					Esters	2
					Acetone	1
Manual cleaning of equipment with solvents	51	3-28	5.7	0.5-30	Xylene	33
					Butanol	8
					Toluene	13
					Dichloromethane	9
					Esters	7
					Ketones	4

^aFrom Ulfvarson (1977)^bSum of ratios of individual solvent levels to their occupational exposure limits^cSolvents constituting at least one-fifth of individual combined exposure levels

The overall improvement in exposure levels over time has been attributed in large part to better control measures and to the increasing production of water-based paints. These results are corroborated by other estimates of the evolution of average solvent exposures in the Swedish paint manufacturing industry with the following values for the overall CEI: 2 in 1950-69, 1.5 in 1970-74, 0.7 in 1975-79 and 0.3 since 1980 (Lundberg, 1986). Heavy naphthas, toluene and benzene are reported to have been the most commonly used solvents during the 1930s, presumably with high exposure levels. Substitutes for aromatic hydrocarbons,

Table 16. Exposure levels (8-h time-weighted average) to organic solvents of 47 paint manufacturing workers^a

Solvent	No. exposed	Exposure (mg/m ³)	
		Median	Range
Xylene	44	82	1-6070
Toluene	43	10	1-1260
Isobutanol	36	4	1-1040
<i>n</i> -Butanol	35	6	1-1540
Ethanol	33	12	1-1090
Ethyl acetate	32	26	1-767
<i>n</i> -Butyl acetate	31	9	1-1680
White spirits	18	44	5-74
Methyl acetate	11	13	3-169
Dichloromethane	5	719	10-2420
Methyl ethyl ketone	5	39	8-124
Isopropanol	3	129	6-258

^aFrom Lundberg & Håkansson (1985)

Table 17. Average combined organic solvent exposure^a of paint industry workers in various work areas over three periods^b

Work area	Period		
	1969 and earlier	1970-75	1976 and later
Industrial paint section			
Mixing	2	1.3	0.7
Grinding	3	1.8	0.9
Tinting-finish	2.2	1.5	0.6
Tapping	2.2	1.2	0.6
Cleaning of vessels	4.5	3	1.5
Alkyd paint section (mixing, tinting, tapping)			0.1-0.2
Small batch manufacturing		1.4	0.7
Filler manufacturing	0.2	0.15	0.1
Storage	0.2	0.15	0.1
Cellulose paint section	2.5		
Laboratory			
Product development	0.7	0.4	0.15
Control laboratory	1	0.75	0.4
Process engineering	2	1	0.4

^aSum of ratios of individual solvent levels to their occupational exposure limits; solvents considered: acetone, butanol, butylacetate, ethanol, ethyl acetate, white spirits, methyl isobutyl ketone, toluene and xylene

^bFrom Ørbaek *et al.* (1985)

including turpentine, decaline and tetraline, would have been used during the Second World War and immediately after. From 1950 until today, the most commonly used solvents would have been xylene, toluene, white spirits, ethanol, butanol, ethyl acetate and butyl acetate. While operations were largely manual before the late 1960s, improvements such as local exhaust ventilation were gradually introduced in the mid-1960s.

(ii) *Exposure to dusts*

In a Swedish investigation covering ten factories manufacturing paint and industrial coatings, dust was found during tinting, handling of bags, compressing empty bags, floor cleaning and emptying air-cleaner filters. The principal exposure to dust, however, was found during charging of raw materials. Sixty-one breathing-zone samples taken over durations of 5 min to 8 h indicated total dust exposure levels of 1.7–70 mg/m³. Raw materials charged included inorganic and organic pigments and fillers, chromium and lead compounds, talc and silica. The highest total dust levels (range, 7.7–70 mg/m³; four samples) were found in a powder coatings factory. Local exhaust ventilation was widely used, but fewer than half of the workers wore respirators. A few air samples were obtained to evaluate exposure to specific dusts during charging operation in some of the companies. Quartz was measured in five factories, with levels ranging from 0.01 to 0.9 mg/m³. Asbestos levels ranged from 0.3 to 5 fibres/cm³ (four factories). Chromium levels (as CrO₃) ranged from 0.003 to 1.6 mg/m³ (seven factories), while lead levels ranged from 0.006 to 4 mg/m³ (three factories; Ulfvarson, 1977). Blood lead concentrations monitored in 80 workers in 12 paint manufacturing companies in Finland were 5–72 µg/100 ml. The highest value was found in a spray painter (Tola *et al.*, 1976).

(iii) *Other exposures*

Exposure to ammonia was reported while charging it for use in water-based paints in the Swedish paint industry, at average levels of 50–80 ppm (35–56 mg/m³). In one case, more than 700 ppm (490 mg/m³) was measured. The levels of pentachlorophenol and phthalic anhydride were below the standards of 0.5 mg/m³ and 2 ppm (12 mg/m³), respectively (Ulfvarson, 1977). The concentration of diethylene triamine was below the detection limit (0.01 mg/m³) in the breathing zone of two workers canning epoxy paint curing agents in a Finnish paint factory (Bäck & Saarinen, 1986).

In a US paint manufacturing company, the 8-h time-weighted average (TWA) concentration of vinyl acetate ranged from 1.0 to 8.4 ppm (3.6–30.6 mg/m³; four samples). Personal and area air samples indicated concentrations of ethyl acrylate ranging from below the limit of detection to 5.8 ppm (23.8 mg/m³); concentrations of butyl acetate were all below the limit of detection (16 samples), except one sample at 0.9 ppm (4.7 mg/m³; Belanger & Coye, 1980).

(c) *Construction painting and lacquering*

Usual painters' work in the construction industry involves the use of a rather limited number of types of coatings – mainly decorative water- or solvent-based paints and wood lacquers and varnishes. The potential for exposure to a variety of substances (mainly solvents and pigments) is high, however: painting performed inside buildings, where poor ventilation opportunities, especially in confined spaces such as small rooms, cupboards, bath-

rooms, can lead to very high levels of contaminants; whereas when painting the outside of buildings (facades, windows, roofs), natural ventilation is usually effective. Painting of new buildings usually involves mainly water-based paints and spraying equipment; however, during renovation or maintenance, solvent-based paints are still widely used and work is usually performed by hand with a brush or roller.

Surfaces to be coated can be made of plaster- or gypsum-based wall-board composite materials, concrete, wood such as for windows, doors and flooring, and more rarely metal. Construction painters may spend a good proportion of their time in preparatory or accessory work. In a Finnish study on construction painters, 92 of 231 (40%) painters estimated that they spent more time on such work than actually painting (Riala *et al.*, 1984). Removing old paint and preparing surfaces in general may involve the use of paint strippers containing solvents such as dichloromethane, of gas-operated blow torch units or hot air guns which may generate organic pyrolysis fumes, metallic fumes and dusts from pigments containing *inter alia* chromium, lead and arsenic compounds. Other accessory tasks may be polishing, sanding or sandblasting operations, which generate old paint, quartz, concrete, plaster, wood and metal dusts. Acid or alkali washing solutions may be used, as well as steam generators for removing wallpaper, which release carbon monoxide-containing exhaust gases. Preparing surfaces also often involves filling cracks and holes using plaster, cement, sealers, spackling, taping and dry wall materials, putties and wood fillers, implying possible additional exposure to inorganic dusts and fibres (including asbestos) and solvents. Further exposure stems from the use of solvents during the cleaning of equipment as well as for personal cleaning (Ringen, 1982; Huré, 1986; Swedish Work Environment Fund, 1987).

The use of solvents in construction paints, and thus painters' exposures, has evolved radically with time. Early whitewashes and distempers contained no organic solvent, and oil paints contained only about 10% turpentine or, later, white spirits. Alkyd paints introduced in the 1960s required approximately 50% of a solvent such as white spirits. With the introduction of epoxy paints for special surfaces such as floors, other solvents such as alcohols, esters and aromatic hydrocarbons became more widely used. Water-based latex paints were introduced in the 1950s but were more widely accepted in the 1960s and 1970s, to become predominant in the 1980s; now, an estimated 60–80% of building trade coatings are water-based (Dufva, 1982; Hansen, 1982; Riala *et al.*, 1984). Vinylic and acrylic water-based paints are the most common, and these contain only a small percentage of organic solvents, mainly alcohols or glycol ethers.

The 8-h TWA exposure to solvents of 45 Dutch maintenance painters working on 12 different projects has been measured. Summed air concentrations averaged 101 mg/m³ (geometric mean) for the whole group and 59 mg/m³ for a subgroup of 20 house painters who applied only alkyd resins by brush and roller. Benzene was detected at only two of the sites and at low concentrations (up to 0.2 mg/m³). Toluene concentrations were below 4 mg/m³, except at one site where it reached 43 mg/m³. C₂- and C₃-substituted benzenes and C₈-C₁₁ alkanes were found at most sites, originating mainly from the use of white spirits. Workers using chlororubber paint in a pumping station were exposed to carbon tetrachloride at levels

ranging from 10 to 17 mg/m³; the highest level of toluene was also found at this site (Scheffers *et al.*, 1985).

The exposure of Danish house painters to 13 solvents was investigated in 1974. Overall exposure, standardized to relevant occupational exposure limits, was above the permissible limit for five of 11 maximal values, reaching up to 34 times the permissible limit. Individual solvent average exposure levels were especially elevated for benzene (55 ppm [175 mg/m³]; 41 samples), believed to originate from thinners, and for trichloroethylene (91 ppm [490 mg/m³]; 33 samples). The origin of the trichloroethylene was not specified (Mølhav & Lajer, 1976).

In Finland, concentrations of Stoddard solvent during application of solvent-containing alkyd paints were 22–65 ppm (seven samples) and those during application of wood preservatives or alkyd varnishes, 68–280 (four samples). The overall solvent CEI during parquet floor varnishing using cellulose nitrate lacquers and urea-formaldehyde varnish ranged from 0.6 to 2.3, according to Finnish occupational exposure limits. Acetone, ethanol, isobutanol and butyl acetate were the main solvents used. Exposure to formaldehyde during varnishing averaged 2.8–4.5 ppm (3.4–5.5 mg/m³; Riala, 1982). The risk of formation of bis(chloromethyl)ether (see IARC, 1987s) from the reaction between formaldehyde and hydrochloric acid (used as a hardener) in urea-formaldehyde varnishes has been evoked (Dufva, 1982), but levels higher than 0.2 ppb (>0.9 µg/m³) have not been found (O'Neill, 1981).

In a study in Finland mainly of maintenance construction workers, the overall average airborne concentration of solvents during alkyd and urethane painting and varnishing, expressed as solvent naphtha exposure, was 132 ppm (77 samples); this was much higher when there was no ventilation, either natural or artificial (197 ppm; 46 samples), than with ventilation (38 ppm; 31 samples). Highest concentrations were observed during painting in small, unventilated rooms (303 ppm) and on large surfaces such as walls and ceilings with no ventilation (206 ppm with roller and brush painting and 243 ppm with spray painting). Taking into account other activities, e.g., use of water-based paints, the overall average 8-h TWA exposure level was 40 ppm (Riala *et al.*, 1984).

Air concentrations (mg/m³) of organic vapours generated during the application of water-based paints were measured by personal sampling in Denmark, as follows: butyl acrylate, 0–2; diethylene glycol butyl ether, 4–5; diethylene glycol methyl ether, 8–32; dipropylene glycol methyl ether, 30–40; ethylene glycol butyl ether, 2–60; ethylene glycol phenyl ether, 0–0.7; propylene glycol, 2–70; 2,2,4-trimethylpentane-1,3 diol monoisobutyrate, 0.5–12; triethylamine, 4–6; and white spirits, 40–75. Concentrations of two gases, formaldehyde (at 0–0.4 mg/m³) and ammonia (at 2–12 mg/m³) were also reported (Hansen *et al.*, 1987).

In a Swedish study of renovation spray painters, very high concentrations of white spirits (1200–1500 ppm) were measured during use of alkyd-type paints and 100–1000-times lower concentrations of solvents during use of acrylate-polyvinyl acetate-based water-borne paints. Dust concentrations, originating from paint mist, were higher during use of water-based paints (77–110 mg/m³) than solvent-based paints (17–27 mg/m³). Inorganic substances were found to represent 80% and 70–85% of the dust content, respectively. Exposures to

substances such as lead (in solvent-based paints) and zinc (in both types of paints) were 10–23% and 1–2% of their respective exposure limits (Bobjer & Knave, 1977).

The mean blood lead level measured in 1962 for a group of 107 decorative and house painters in the USA was 23 $\mu\text{g}/100\text{ g}$ blood, similar to that in control groups (Siegel, 1963).

(d) *Painting, varnishing and lacquering in the wood industry*

Application of clear varnish or lacquer finishes on furniture represents the main use of coatings in the wood industry. Paints, varnishes and lacquers are also used in the production of various wooden raw materials (e.g., composite wood boards) and miscellaneous wooden articles (e.g., toys, tableware). Until the mid-1950s, cellulose ester-type lacquers were almost the only ones used in the furniture industry; however, amino resin-based, polyurethane and polyester coatings now constitute the main coatings in the industry (Swedish Work Environment Fund, 1987).

Workers are exposed mainly through inhalation or cutaneous absorption of solvents either from paint mist or from vapours generated by spraying operations, from vapours evolved from finished products or from auxiliary work such as mixing the coatings, cleaning equipment or applying other products such as wood fillers and sealants. The amount of exposure is influenced by the method of applying coatings; the most common are spraying, usually at low pressure, curtain and roller coating and dipping. The main categories of solvents used are aliphatic esters, ketones, alcohols and hydrocarbons as well as aromatic hydrocarbons (O'Brien & Hurley, 1981; Swedish Work Environment Fund, 1987).

Low molecular-weight resin constituents such as formaldehyde and isocyanates may be evolved during application or curing of coatings. Another possible exposure is to wood dust from the general factory environment and from preparatory work such as sanding.

Air monitoring was carried out over a ten-year period (1975–84) in 50 Finnish furniture factories, where the main coatings used were acid-cured amino resin-based paints and varnishes. The most commonly used solvents were xylene, *n*-butanol, toluene, ethanol, butylacetate and ethylacetate, which were present in more than 50% of 394 measurements. Mean concentrations of the solvents present were below 20 ppm, except for white spirits, which occurred at 66 ppm. Arithmetic mean solvent vapour concentrations measured during different work tasks ranged from 0.4 ppm in spray painting to 2.1 ppm during cleaning of a painting machine, with individual values varying from 0.1 to 7.4 ppm. Formaldehyde, derived from the amino resin binder, was the object of 161 short-term measurements (15–30 min) covering different work tasks. The arithmetic mean of the concentrations varied from 0.9 to 1.5 ppm (1.1–1.8 mg/m^3), with individual values ranging from 0.1 to 6.1 ppm (1.2–7.5 mg/m^3 ; Priha *et al.*, 1986).

The 8-h TWA exposure to formaldehyde of 38 employees in a Swedish light furniture factory applying acid-hardening clear varnishes and paints was found to average 0.4 mg/m^3 (range, 0.1–1.3) with a mean exposure to peak values (15 min) of 0.7 mg/m^3 . Mean exposure to solvents was low. The dust concentration was low – usually less than one-tenth of the Swedish threshold limit value of 5 mg/m^3 (Alexandersson & Hedenstierna, 1988).

In a study of a US wood furniture company producing stereo equipment cabinets, the solvent exposure of 27 employees in spray painting and finish wiping operations was mea-

sured. In spraying jobs that involved the use of an acrylic base coat, an oil-based glaze or stain and cellulose nitrate lacquers, total exposure to paint mist (8-h TWA) varied from 0.1 to 2.5 mg/m³ (geometric means). Combined exposure to solvents (CEI) varied from 0.05 to 0.11 in base coat operations (solvents measured: methyl ethyl ketone, isopropyl acetate, xylene, isopropanol, methyl isobutyl ketone, toluene and isobutyl isobutyrate), from 0.06 to 0.10 in glaze operations (toluene, xylene, ethylene glycol monobutyl ether and petroleum distillates) and from 0.08 to 0.24 in lacquer operations (isopropanol, ethanol, isophorone, isobutyl acetate, *n*-butanol, toluene, xylene, ethylene glycol monobutyl ether, methyl ethyl ketone, isobutyl isobutyrate, isopropyl acetate and petroleum distillates). The overall low air concentrations of paint mist and organic solvents were attributed to adequate ventilation in paint booths and good working practices (O'Brien & Hurley, 1981).

Exposure to organic solvent vapours was also measured in 16 small-scale industries in Japan, where synthetic *urushi* lacquer was applied to wooden tableware (bowls), vases and altars, and in two furniture factories. Work involved mainly brush painting, screen painting and hand-spraying operations. Toluene, xylene, ethylbenzene and *n*-hexane were the recorded solvents. Average mixed solvent personal exposure (CEI) was always low (below 0.44), except in the case of an automated spray operation (1.4; Ikeda *et al.*, 1985).

The average 4-h exposure to toluene of 20 workers employed in painting and hand-finishing in an Italian art furniture factory was 27–182 mg/m³. Toluene was the principal solvent found in the work environment; other major solvents found were acetone, isobutanol, ethanol and ethyl acetate (Apostoli *et al.*, 1982).

In the Finnish plywood industry, solvent concentrations in workroom air were recorded during coating operations involving polyurethane and alkyd paints. The following ranges in ppm (mg/m³) were obtained from eight to 12 measurements: (i) polyurethane paint: methyl isobutyl ketone, 2–28 (8.2–115); butylacetate, 8–50 (38–238); xylene, 10–25 (43–108); and cyclohexane, 1–28 (3.4–95); (ii) alkyd paint: toluene, 2–3 (7.5–11.3); xylene, 7–12 (30.4–52); isobutanol, 7–11 (21–33); and trimethylbenzene, 1–9 (5–44; Kauppinen, 1986).

In a US plant where paint was stripped from wood and metal, breathing zone TWA concentrations of dichloromethane for three operators ranged from 633 to 1017 mg/m³ in seven samples (Chrostek, 1980).

(e) *Painting in the metal industry*

Protection from corrosion is the primary aim of metal painting. Mild steel is thus almost always subjected to the application of a primer coat containing corrosion inhibitors such as iron and lead oxides or of zinc powder, further covered with a decorative paint. Aluminium may be covered with a zinc chromate-based primer before a decorative coat is applied.

During the preparation of metal parts, painters may be exposed to cleaning and degreasing agents, such as solvents, alkalis and acids, and to abrasive dusts, such as crystalline silica generated during blast cleaning. Depending on the industry, metal painters may be exposed to a variety of dusts, solvents, fumes and gases resulting from operations such as mixing paints, maintaining equipment, applying fillers, sealers or putty, or background metal welding or assembling operations. Most coatings used in the metal industry are solvent-based, and spray painting is the main method of application, leading to potential exposures to

paint mist and solvents. Two-component paints, such as those based on epoxy and polyurethane resins, play a major role, implying potential exposure to reactive substances such as isocyanates and epoxides. Air-drying or baking after application results in the evolution of solvents and, possibly, thermal degradation products of resins (Peterson, 1984).

(i) *Exposure to organic solvents*

Exposure of metal spray painters to a variety of solvents has been measured by the US National Institute for Occupational Safety and Health in a number of industries. The results are summarized in Table 18. Except in railroad car painting, overall exposure levels were found to be low. Toluene, xylene and petroleum distillates were among the most common solvents. Analyses of bulk air samples indicated no detectable benzene (O'Brien & Hurley, 1981).

In Finland, solvent concentrations were measured in the breathing zone of 40 car painters at six garages (54 1-h samples). Mean concentrations and the upper limits of various solvents were as follows (ppm) [mg/m³]: toluene, 30.6 (249) [115 (940)]; xylene, 5.8 (36) [25 (156)]; butylacetate, 6.8 (128) [32 (608)]; white spirits, 4.9 (150); methyl isobutyl ketone, 1.7 (39) [7 (160)]; isopropanol, 2.9 (85) [7 (209)]; ethyl acetate, 2.6 (14) [9 (50)]; acetone, 3.1 (25) [7 (60)]; and ethanol, 2.9 (27) [6 (51)] (Husman, 1980).

A large study of Swedish car refinishing workshops showed that painters spent only 15% of their time actually spray painting, the rest being occupied with grinding, filling, masking and assembling activities (60%) and colour mixing, degreasing and cleaning activities (25%). The highest overall solvent exposure was observed during spray painting, with a combined exposure of 0.3 (CEI; 106 samples). Toluene, xylene and ethyl acetate were present in all samples, at average levels of 39, 14 and 11 mg/m³, respectively. Ethanol, butanol and butyl acetate were observed at very low levels in nearly half the samples. Other solvents encountered frequently in other activities, although at low levels, included styrene and white spirits. A reconstitution of working conditions in 1955 indicated that exposure levels to solvents were higher than in 1975, which was considered to be representative of the 1960s and 1970s. In particular, when benzene was used as a solvent in 1975–77, the combined exposure (CEI) reached 0.8 (Elofsson *et al.*, 1980).

Breathing zone samples were taken during short-term spray painting operations in a small autobody repair shop in the USA. Elevated levels of total hydrocarbons (up to 1400 ppm) were measured in winter when the spraybooth fan was turned off to conserve heat. Under these conditions, high concentrations of toluene (590 ppm; 2224 mg/m³) were seen during lacquer spray painting and of xylene (230 ppm; 1000 mg/m³) and benzene (11 ppm; 35 mg/m³) during enamel spray painting. Summer conditions, when the fan was on, resulted in maximal concentrations of 330 ppm total hydrocarbons, 56 ppm (211 mg/m³) toluene, 44 ppm (191 mg/m³) xylene and 3.7 ppm (12 mg/m³) benzene. Other major solvents measured were acetone, cellosolve acetate, methyl isobutyl ketone, *n*-hexane, methyl cellosolve acetate, trimethylbenzene, ethylbenzene and *n*-butyl acetate (Jayjock & Levin, 1984).

Table 18. Painters' time-weighted average exposure levels (personal breathing-zone samples) in various metal spray-painting operations^a

Operation	Sampling time	No. of samples	Combined exposure ^{b,c}	Main solvents measured	Concentration (mg/m ³) ^c
Light aircraft finishing, primer spraying	25-41 min	3	0.9 ± 1.5	2-Butanone	42 ± 2.1
				Toluene	60 ± 1.2
				Ethanol	26 ± 1.6
				Isopropanol	19 ± 1.6
Light aircraft finishing, topcoat spraying	27-62 min	7	0.15 ± 1.3	Ethylacetate	77 ± 1.3
				Ethoxyethylacetate	44 ± 1.4
				Aliphatic hydrocarbons	34 ± 1.2
Light aircraft finishing, stripping operations	19-35 min	6	0.13 ± 2.5	Ethylacetate	52 ± 2.5
				Ethoxyethylacetate	30 ± 2.7
				Aliphatic hydrocarbons	73 ± 1.5
Car refinishing	15-45 min	7	0.09 ± 1.5	Toluene	39 ± 1.6
				Xylene	10 ± 1.0
				Petroleum distillates	21-63
				Other solvents	< 10
Railroad car	15-60 min	14	1.3 ± 1.4	Toluene	188 ± 1.5
				Xylene	14 ± 2.6
				Other aromatic compounds	217 ± 1.4
				Aliphatic hydrocarbons	840 ± 1.4
Heavy equipment	60 min	12	0.01-0.05	Refined solvents	21-96
				Other solvents	≤ 5
Metal furniture, solvent and water-borne paints	8 h	5 painters	0.10-0.46	Toluene	12-61
				Xylene	7-48
				<i>n</i> -Butyl acetate	22-109
				Diisobutyl ketone	< 1-23
				2-Ethoxyethyl acetate	1-14
				Aliphatic hydrocarbons	33-180
Metal furniture, high-solids paints	8 h	6 painters	0.07-0.31	Xylene	6-55
				Aromatic distillates	5-60
				Other solvents	< 10
Appliance finishing	8 h	4 painters	0.38-0.79	Toluene	88-204
				Xylene	112-225

^aFrom O'Brien & Hurley (1981)^bCumulative exposure index (see p. 366), based on US Occupational Safety and Health Administration permissible exposure levels^cGeometric means ± geometric standard deviation, unless otherwise stated

In Japan, the full-shift TWA concentrations of the following solvents measured for 13 car repair painters (ppm [mg/m^3]; mean \pm standard deviation) were: xylene, 8 ± 8 [35 ± 35]; toluene, 19 ± 13 [72 ± 49]; isobutanol, 5 ± 5 [15 ± 15]; and ethyl acetate, 6 ± 4 [22 ± 14]. The overall combined exposure (CEI) was 0.38 ± 0.25 . Short-term samples taken during painting showed a higher combined exposure for ten of 14 workers, toluene being the major solvent encountered (Takeuchi *et al.*, 1982). In another Japanese study of car refinishing painters, high toluene concentrations were observed during painting in side-wall ventilated booths (410–660 ppm; 1546–2488 mg/m^3), compared with those in downdraft ventilated booths (28–87 ppm; 106–328 mg/m^3). Short actual painting periods resulted in full-shift TWA concentrations of organic solvents (toluene, xylene, methyl acetate, ethyl acetate and butyl acetate) below the exposure limits. The average hippuric acid concentration in the urine of painters (0.33 mg/ml) was slightly higher than that in controls (0.19 mg/ml ; Matsunaga *et al.*, 1983).

Exposure to toluene was investigated in 1940–41 in 106 painters in a large US airplane factory. Eight-hour TWA levels of toluene ranged from 100 to 1100 ppm (377–4147 mg/m^3); approximately 60% of workers were exposed to 200 ppm (754 mg/m^3) or more. Toluene was a major constituent of zinc chromate primers, lacquers, cellulose nitrate dope (lacquer) and brush wash (Greenburg *et al.*, 1942).

An industrial hygiene evaluation was conducted at a commercial airline maintenance facility in the USA. Employees working in and around jet aircraft during the paint stripping process were exposed to levels of dichloromethane in the breathing zone that ranged from 79 to 950 mg/m^3 with a mean of 393 mg/m^3 . During application of the prime coat, exposure to solvents was as follows (mg/m^3 ; mean and range): toluene, 112 (51–179); methyl ethyl ketone, 39 (8–77); butyl acetate, 72 (29–130); *n*-butanol, 25 (9–47); isopropanol, 51 (undetectable to 132); and cyclohexanone 10 (undetectable to 23). During application of the top coat, exposure to the solvents was: ethyl acetate, 333 (undetectable to 857); methyl ethyl ketone, 69 (undetectable to 219); methyl isobutyl ketone, 44 (nondetectable to 117); butyl acetate, 80 (undetectable to 210); xylene, 21 (undetectable to 49) and cellosolve acetate, 18 (undetectable to 46; Okawa & Keith, 1977).

Another study involved workers spray painting large commercial aircraft. Industrial hygiene measurements indicated short-term personal exposures as follows (mg/m^3 ; mean and range): toluene, 583 (140–1230); methyl ethyl ketone, 1436 (240–3250); ethyl acetate, 1231 (160–3520); naphtha, 44 (20–120); butyl acetate, 64 (20–150); xylene, 318 (60–1330); cellosolve acetate, 4843 (670–25 170); and dichloromethane, 654 (undetectable to 2840). Long-term exposures to the solvents were: ethyl acetate, 264 (10–1100); methyl ethyl ketone, 197 (20–440); toluene, 162 (30–450); butyl acetate, 11 (undetectable to 50); naphtha, 10 (undetectable to 160); xylene, 69 (10–270); cellosolve acetate, 640 (70–2490); and dichloromethane, 100 (undetectable to 760; Herwin & Thoburn, 1975).

Airborne concentrations (CEI) of solvent mixtures for jobs as paint mixer and spray painter ranged from 0.03 to 0.32 at a US plant manufacturing school and general purpose buses. The solvents found at the plant were petroleum naphtha, toluene, xylene, benzene, methyl ethyl ketone and *n*-hexane (Zey & Aw, 1984).

UK shipyard painters working in ships' accommodation and bilges were exposed to various mean TWA levels of organic solvents, depending on their job: 125 mg/m³ for three painters using a chlorinated rubber paint with white spirits as solvent, 215 mg/m³ for a worker using paint stripper with dichloromethane as the main solvent and 577 mg/m³ for four men using white interior paint with white spirits as the main solvent. Other paint solvents used frequently in dockyards are methyl-*n*-butyl ketone, *n*-butanol, trichloroethylene, xylene and cellosolve (Cherry *et al.*, 1985).

In Poland, phenol and hippuric acids were measured in 51 urine samples from shipyard painters working in small spaces of superstructures and in large holds. The average values of phenol in urine were 12.4–66.4 mg/l compared to 7.9 mg/l on average for a control group. Urinary phenol was attributed to benzene: the benzene concentration in air ranged from undetectable to 11 ppm (35 mg/m³). The average concentrations of hippuric acids in urine (sum of hippuric and methylhippuric acids) were 1812–5500 mg/l compared to 790 mg/l in a control group. Concentrations of toluene and xylene in air were 7–88 ppm (26–332 mg/m³) and 23–538 ppm (100–2335 mg/m³), respectively (Mikulski *et al.*, 1972). Elevated values of hippuric (up to 6700 mg/l) and methylhippuric acids (up to 7100 mg/l) were also measured in the urine of shipyard workers in Japan (Ogata *et al.*, 1971).

In a factory producing dump-truck bodies and earth-moving machinery in the UK, full-shift personal exposure levels to xylene and white spirits (two samples) were measured as 52 and 65 ppm (226 and 282 mg/m³) xylene and 7 and 12 ppm white spirits. After ventilation was properly adjusted, these levels dropped to 9 and 7 ppm (39 and 30 mg/m³) xylene and < 5 ppm white spirits (Bradley & Bodsworth, 1983). At a US plant where truck bodies and refuse handling equipment were manufactured, breathing zone concentrations of xylene during spray painting operations (eight samples varying from 1 to 3 h) ranged from 5 to 140 ppm (22–608 mg/m³; Vandervort & Cromer, 1975). Low exposure levels of toluene (3–18 mg/m³) and isobutyl acetate (2–44 mg/m³) were observed for Swedish spray painters in a plant manufacturing fireplaces (Hellquist *et al.*, 1983).

(ii) *Exposure to paint mists, dusts and specific metals*

Exposures of metal spray painters to paint mists, lead and chromium have been measured by the US National Institute for Occupational Safety and Health in a variety of industries. The results are summarized in Table 19. High concentrations of paint mist have been recorded in several operations, often linked with the painting of enclosed spaces and internal cavities, faulty ventilation and work practices. Substantial but short-term lead exposure was encountered in situations where lead-based pigments were used, such as for painting transportation and heavy equipment. Elevated but brief exposures to chromium were noted during the spraying of aircraft with primer. No antimony, arsenic, cadmium or mercury was encountered in these studies. Very low levels of tin (2–7 µg/m³) were recorded during the spray painting of dibutyltin dilaurate containing enamel on light aircraft (O'Brien & Hurley, 1981).

In a large study of Swedish car refinishing workshops, averages of 7 mg/m³ mist, 100 µg/m³ lead and 26 µg/m³ chromium were measured during spraying activities; during grinding activities, the corresponding values were 3 mg/m³, 20 µg/m³ and 6 µg/m³. The conditions

Table 19. Painters' exposure (personal breathing-zone samples) to paint mists, lead and chromium in various metal spray-painting operations^a

Operation	Sampling conditions	No. of samples	Exposure level ^b		
			Paint mist (mg/m ³)	Lead (µg/m ³)	Total (µg/m ³)
Light aircraft finishing, primer spraying	25-41 min	3	23.3 ± 1.6	ND	1600 ± 1.6
Light aircraft finishing, topcoat spraying	27-62 min	6	23.3 ± 1.7	ND	-
Light aircraft finishing, stripping operations	19-35 min	6	14.1 ± 2.0	ND-5000	-
Car refinishing	15-45 min	7	8.7 ± 1.6	52 ± 1.5	-
Car refinishing	8 h	7	5.0	30	-
Railroad car	15-60 min	13	43.3 ± 1.4	211 ± 1.7	220 ± 2.2
Heavy equipment	60 min	3	2.0-36.5	230-1300	31-230
Metal furniture	8 h	6 painters	3.7-27.6	ND-1050 ^c	-
Metal furniture, high-solids paints	8 h	6 painters	0.5-6.2	5-26	5-9
Small appliance parts, powder coating, electrostatic spraying	8 h	3	1.3 ± 1.1	-	-
Appliance finishing	8 h	4 painters	21.7-54.5	<6-20	-

^aFrom O'Brien & Hurley (1981)

^bGeometric means ± geometric standard deviation, unless otherwise indicated

^cOnly 2.5-h samples taken one afternoon showed detectable levels (30-1050)

were thought to be representative of those in the 1960s and 1970s. Simulation of work conditions in 1955 showed low concentrations of lead during the use of all colours except red, when the Swedish exposure limit was exceeded by 70-fold. The actual exposure of painters was believed to be reduced by the use of individual protective equipment (Elofsson *et al.*, 1980). Breathing-zone samples were taken during short-term spray painting operations in a small auto-body repair workshop in the USA. Only one of eight samples, corresponding to a red paint formula, contained significant levels of chromium (490 µg/m³) and lead (210 µg/m³); in all other measurements, the levels of chromium, lead and cadmium were below the detection limit. The concentration of total dust collected during the sanding or grinding of plastic body filler was 5-40 mg/m³ and that of respirable dust, 0.3-1.2 mg/m³ (Jayjock & Levin, 1984). In a factory producing dump-truck bodies and earth-moving machinery in the UK, full-shift personal exposure levels to total paint solids (two samples) were measured as 11.6 and 15.9 mg/m³. After ventilation was properly adjusted, these levels dropped to 1.4 and 5.2 mg/m³. The major pigments used were titanium dioxide and iron oxide (Bradley & Bods-

worth, 1983). At a US plant where truck bodies and refuse handling equipment were made, breathing zone concentrations of solid contaminants measured during various spray painting operations (seven samples varying from 1 to 3 h) were 4.8–47 mg/m³ for total particulates, 20–3000 µg/m³ lead and 10–400 µg/m³ chromium (Vandervort & Cromer, 1975). Low overall exposure levels were found for Swedish spray painters working in a plant manufacturing fireplaces, with a total dust level of 1.7 mg/m³, chromium oxide, 5–8 µg/m³ and zinc oxide, 20–30 µg/m³ (Hellquist *et al.*, 1983).

At a US plant where school and general purpose buses were manufactured, employees working in and around the paint booth were reported to be exposed to hexavalent chromium and lead. The concentrations of hexavalent chromium in five personal air samples were 0.03–0.45 mg/m³, with a mean of 0.23 mg/m³; airborne lead concentrations (eight personal samples) ranged from below the laboratory limit of detection (3 µg/filter) to 2.01 mg/m³, with a mean of 0.78 mg/m³ (Zey & Aw, 1984).

A US manufacturer of large-scale weapon, electronic and aero-mechanical systems reported exposure of workers to hexavalent chromium while spraying aircraft wheels with yellow lacquer primers containing zinc chromate. In 12 personal breathing zone samples, the level of chromium[VI] ranged from 13.3 to 2900 µg/m³ with a mean of 606.7 µg/m³ (Kominisky *et al.*, 1978).

In a US plant in which bridge girders were sprayed with lead silico-chromate paint, personal air levels of lead and chromium (as Cr) in five samples were 0.01–0.25 mg/m³ (mean, 0.08) and 0.01–0.04 mg/m³ (mean, 0.02), respectively (Rosensteel, 1974). Substantial exposure to airborne lead was demonstrated for US workers involved in scraping old lead-based paint from the metallic structure of a bridge and priming it (24–1017 µg/m³); recoating with lead-based paint implied lower exposure levels (6–30 µg/m³). Blood lead levels in these workers were 30–96 µg/100 ml, with 58% above 60 µg/100 ml (Landrigan *et al.*, 1982). In the Netherlands, workers involved in flame-torch cutting of a steel structure coated with lead-based paints were shown to be exposed to 2–38 mg/m³ airborne lead (Spee & Zwennis, 1987).

Blood lead levels have also been measured in workers in various occupations in three Finnish shipyards. Painters were among the most heavily exposed: mean blood levels in a total of 77 painters in the three shipyards were 20–28 µg/100 ml (Tola & Karskela, 1976).

(iii) Other exposures

Use of polyurethane type paints can result in exposure to diisocyanate monomers and their oligomers. In Sweden, 43 car repair painters were exposed to a TWA of 115 µg/m³ hexamethylene diisocyanate (HDI)-biuret oligomer, with a range of 10–385 µg/m³. Very high exposure peaks (up to 13 500 µg/m³) were measured. The concentration of HDI was 1.0 µg/m³ (Alexandersson *et al.*, 1987). In Finland, average HDI and HDI-biuret oligomer levels in four car paint shops during spray painting (ten 5–10-min personal samples, outside respirator) were 49 (± 22 SD) and 1440 (± 1130) µg/m³, respectively. The proper use of a respirator with combined charcoal and particle filters was shown to reduce exposure levels to below detection limits (Rosenberg & Tuomi, 1984). In a US car repainting shop, three short-term air samples (5–13 min) taken in the breathing zone during spray painting operations showed HDI levels of < 130 µg/m³. Similar measurements taken during various light air-

craft finishing operations (7–21 min, eight samples) indicated HDI levels below approximately $70 \mu\text{g}/\text{m}^3$, except for one operation with a level of $250 \mu\text{g}/\text{m}^3$ (O'Brien & Hurley, 1981).

Ambient levels of HDI during the spray application of an enamel top coat at a US airline maintenance facility were $< 0.04\text{--}3.20 \text{ mg}/\text{m}^3$, with a mean of $1.1 \text{ mg}/\text{m}^3$ (Okawa & Keith, 1977).

Epoxy paints are usually applied as reactive mixtures of epoxy resins and curing agents, leading to potential exposure to compounds containing the epoxide group. Total epoxide concentrations have been measured in area samples of aerosols collected during three painting operations involving the use of a bisphenol-A diglycidyl ether type of epoxy resin. In a facility producing military aircraft, the use of an epoxy primer did not result in detectable epoxide levels, and the authors surmised that the epoxy-amine curing reaction had probably consumed most of the epoxide group. Epoxide levels of $2\text{--}12 \mu\text{Eq}/\text{m}^3$ epoxide functional group were recorded during the painting of a tank with coal-tar epoxy coatings and the painting of a metal ceiling using an epoxy architectural coating (Herrick *et al.*, 1988). In a US company that finished structural steel members and other fabricated steel products, the products are blasted with steel shot or sand and spray-painted with two-component epoxy paints or oil-based paints. Personal air levels of epichlorohydrin were reported to range from 2.4 to $138.9 \text{ mg}/\text{m}^3$, with a mean TWA of $64.9 \text{ mg}/\text{m}^3$. Bisphenol A glycidyl ethers were also detected in the workers' breathing zone at levels which ranged from below the limit of detection ($0.6 \mu\text{g}$) to $28.6 \mu\text{g}/\text{m}^3$, with a mean of $9.8 \mu\text{g}/\text{m}^3$ (Chrostek & Levine, 1981).

The major thermal degradation components of epoxy powder paints were identified as phenol, cresols, bisphenol-A, pyridine, 2,3-dimethylpyrazine and formaldehyde; bisphenol-A glycidyl ether was not observed. Levels in the work environment of painters were not measured (Peltonen, 1986; Peltonen *et al.*, 1986). Diethylene triamine, which is a component of curing agents of epoxy paints, was measured in three samples collected from the breathing zone of a painter during spray painting of paper machine cylinders and pulp tanks at a concentration of $0.02\text{--}0.07 \text{ mg}/\text{m}^3$ (Bäck & Saarinen, 1986).

Operators working in eight plants where coal-tar enamel protective coating was applied to pipelines with heat were exposed to high levels of coal-tar pitch volatiles (see IARC, 1985b) at up to $24 \text{ mg}/\text{m}^3$ of benzene-soluble matter (full-shift samples). The overall respirable concentration of benzo[*a*]pyrene in the plants averaged $133 \mu\text{g}/\text{m}^3$ (Larson, 1978).

3. Biological Data Relevant to the Evaluation of Carcinogenic Risk to Humans

3.1 Carcinogenicity studies in animals

No data were available to the Working Group.

3.2 Other relevant data in humans

(a) Toxic effects

(i) Skin and eyes

Workers in the paint manufacturing industry (Pirilä, 1947; Ulfvarson, 1977) and painters (Pirilä, 1947; Högberg & Wahlberg, 1980; Winchester & Madjar, 1986) are at a considerable risk of developing an occupational dermatosis. In one study of Swedish paint industry workers, the prevalence of occupational dermatoses was about 40%, 26% of which were on the hands and arms (Ulfvarson, 1977). Among Swedish house painters, the prevalence of occupational skin disease was 4–6%, mainly affecting the hands. More than half of the dermatoses were nonallergic contact eczemas, probably mostly induced by organic solvents, mainly in atopic subjects. Allergic contact eczemas involved hypersensitivity towards chromium, nickel, epoxy resin components and formaldehyde. Several cases were seen of allergy to chloracetamide, which was widely used as a biocide in water-based paints and glues. Sensitivity to turpentine, which was formerly prevalent among painters, is now rare (Högberg & Wahlberg, 1980).

The wide variety of skin sensitizing agents in paints include some of the monomer residues from resins (e.g., phenol/formaldehyde resins, carbamide resin, melamine resin, epoxy compounds, acrylates). In addition, natural resins, such as colophony, may contain sensitizing agents. Some hardeners, such as acid anhydrides and *para*-toluenesulfonic acid, may cause sensitization, as may some metals used as pigments and driers in paints, e.g., cobalt and zirconium. Chromate sensitivity is rare in the painting trades, due to the low solubility of the salts used. Of other additives, several biocides (e.g., formaldehyde, chlorophenols and isothiazolinones) may have this effect. Of the solvents, only turpentine and dipentene (limonene) are known to be sensitizers (Fregert, 1981; Hansen *et al.*, 1987).

Some organic solvents (e.g. some ketones and esters) are irritants, as are some resin monomers (e.g., butyl acrylate) and additives (e.g., amines, ammonia and organic peroxides; Hansen *et al.*, 1987).

Corneal changes have been described in workers exposed to spray paints containing xylene (Matthäus, 1964). Changes in the lens of the eye have been recorded in car painters exposed to a mixture of solvents (Raitta *et al.*, 1976; Elofsson *et al.*, 1980). However, no ocular effect was noted in industrial spray painters occasionally exposed to toluene at up to 4125 mg/m³ (Greenburg *et al.*, 1942). Water-based paints may contain triethylamine (Hansen *et al.*, 1987), which can cause corneal oedema (Åkesson *et al.*, 1985, 1986).

(ii) Respiratory tract

Complaints of irritation in the upper airways were reported among paint factory workers (Winchester & Madjar, 1986) and among painters occupationally exposed to white spirits and other solvents (Cohen, 1974; Seppäläinen & Lindström, 1982; Lindström & Wickström, 1983; Pham *et al.*, 1985; White & Baker, 1988). Hyposmia has sometimes been associated with exposure of painters to solvents (Lindström & Wickström, 1983). Histological changes of the nasal mucosa were reported among industrial spray painters (Hellquist *et al.*, 1983).

Some painters suffer from lower airway symptoms (Schwartz & Baker, 1988; White & Baker, 1988), and there is a high prevalence of chronic phlegm bronchitis among spray painters (White & Baker, 1988) and lacquerers (Sabroe & Olsen, 1979). An obstructive ventilatory pattern was recorded after testing lung function in people who abused spray paint by inhalation (Reyes de la Rocha *et al.*, 1987). A decrease in expiratory flow rates was noted in a few workers in a printing paint factory, probably due to irritant effects, but not among car painters (Beving *et al.*, 1984a). Other studies of painters have also indicated bronchial obstruction (Pham *et al.*, 1985; Schwartz & Baker, 1988; White & Baker, 1988), and small airways disease has been noted in car painters exposed to isocyanates (Alexandersson *et al.*, 1987). In contrast, no disturbance of lung function was reported among house painters using solvent-based (Hane *et al.*, 1977; Askergrén *et al.*, 1988) and water-based (Askergrén *et al.*, 1988) paints. Danish painters were reported to have a high rate of disability pensions due to respiratory disease (Mikkelsen, 1980).

Painting may also entail exposure to compounds that cause allergic reactions in the airways. Isocyanates can cause both asthma and pneumonitis in painters (Nielsen *et al.*, 1985; Hagmar *et al.*, 1987). Exposure in the painting trade to isocyanates and polyisocyanates may induce antibody formation (Welinder *et al.*, 1988). Acid anhydrides (e.g., trimellitic anhydride, phthalic anhydride and its derivatives, and maleic anhydride) caused sensitization in workers producing alkyd binders (Wernfors *et al.*, 1986; Hagmar *et al.*, 1987; Nielsen *et al.*, 1988). Moreover, paints sometimes contain asthma-inducing amines (Hagmar *et al.*, 1987).

Exposure to aluminium dust and iron oxide during paint production may cause fibrosis, and exposure to iron oxide can cause pneumoconiosis (Maintz & Werner, 1988).

(iii) *Nervous system*

The neurotoxic effects of exposures to solvents have been reviewed (World Health Organization, 1985; Cranmer & Golberg, 1986; National Institute for Occupational Safety and Health, 1987). Such effects have been determined by means of questionnaires about subjective symptoms, neuropsychological testing and neurophysiological examination of central and peripheral nervous system function (Table 20), as well as in epidemiological studies of neuropsychiatric diseases.

Subjective symptoms, e.g., a feeling of intoxication, fatigue, poor concentration, emotional instability, short-term memory problems and headache, have been recorded in a series of cross-sectional studies of workers in the paint manufacturing industry, of house painters, of car and industry painters and of shipyard painters. Some of the symptoms are short or mid-term, others are persistent. However, no such symptom was recorded in house painters using mainly water-based paints (Askergrén *et al.*, 1988). Neuropsychological tests have documented impairment of psychomotor performance, memory and other intellectual functions, as well as changes of mood (Table 20).

Electroencephalographic changes and a slight decrease in cerebral blood flow were recorded in paint industry workers (Ørbaek *et al.*, 1985). Electroencephalographic abnormalities have also been seen in car and industry painters (Seppäläinen *et al.*, 1978; Eloffsson *et al.*, 1980). Other studies of solvent-exposed painters have failed to identify such effects (Seppäläinen & Lindström, 1982; Triebig *et al.*, 1988), and no effect on auditory-provoked

Table 20. Symptoms and neurobehavioural effects in studies of workers in the painting trade^a

Population	Symptoms	Psychomotor performance	Short-term memory	Other intellectual functions	Mood	Reference
Paint industry workers	+	+	+			Anshelm Olson (1982)
	++ [+]	- [+]	-	[+]	++	Ørbaek <i>et al.</i> (1985) Winchester & Madjar (1986)
House painters	[+]	+	+	+		Hane <i>et al.</i> (1977)
	+	++	+	-	-	Lindström & Wickström (1983)
	[++]	[-]	[-]	[-]	[-]	Fidler <i>et al.</i> (1987)
	-	-	-	-	+	Triebig <i>et al.</i> (1988)
	++	[++] ++	[++] [+]	[++] [+]	[++] [++]	Baker <i>et al.</i> (1988) Mikkelsen <i>et al.</i> (1988)
Car/industry painters		+	+	+	+	Hänninen <i>et al.</i> (1976)
	+					Husman (1980)
	+	+	+	+	+	Elofsson <i>et al.</i> (1980)
	+				+	Struwe <i>et al.</i> (1980)
		[-]	[+]	[-]		Maizlish <i>et al.</i> (1985)
Shipyard painters	+	[+]	-	-		Cherry <i>et al.</i> (1985)
	[+]			+		Valciukas <i>et al.</i> (1985)

^a+, exposed group differed statistically significantly from a control group; ++, there was a dose-response relationship; -, there was no statistically significant difference; [], the Working Group considered that the evidence was limited because the effect was weak or inconsistent and/or the duration and/or intensity of the exposure was low.

potential was seen in painters exposed to water-based paints (Askergren *et al.*, 1988). In one group of house painters (Mikkelsen *et al.*, 1988) and in a study of car and industrial painters (Elofsson *et al.*, 1980), signs of slight atrophy were found by computed brain tomography, but another study showed no such effect (Triebig *et al.*, 1988).

Occasional cases of clinical polyneuropathy have been described in spray painters exposed to methyl-*n*-butyl ketone (Mallov, 1976). In a few cross-sectional studies of car and industry painters (Elofsson *et al.*, 1980; Husman, 1980; Maizlish *et al.*, 1985), signs of slight neurological impairment were observed during physical examinations. Neurophysiological studies of house painters (Askergren *et al.*, 1988) and of car and industrial painters (Seppäläinen *et al.*, 1978) have indicated slight toxic effects on the peripheral nervous system, but other studies have not (Seppäläinen & Lindström, 1982; Cherry *et al.*, 1985; Ørbaek *et al.*, 1985;

Triebig *et al.*, 1988). Formerly, painters exposed to lead sometimes showed clinical effects on the peripheral nervous system, including palsy (mainly affecting the extensor muscles of the forearm) and drop (affecting the wrist; Rosen, 1953). No effect on the peripheral nervous system was observed in painters who used mainly water-based paints (Askergren *et al.*, 1988).

In a cohort of Danish painters, statistically significant two- to three-fold increases in the relative risk of being granted a disability pension due to neuropsychiatric disease was found (Mikkelsen, 1980). Similarly increased risks were observed in case-control studies of applicants for disability pensions due to neuropsychiatric disease and for nursing home accommodation due to encephalopathy, in which the occupation of 'painter or other solvent-exposed trade' was used as an indicator of exposure (Axelson *et al.*, 1976; Olsen & Sabroe, 1980; Lindström *et al.*, 1984; Rasmussen *et al.*, 1985). Significant increases in risk were not, however, seen in other case-control studies, using subjects granted a disability pension (van Vliet *et al.*, 1987), subjects who had consulted general practitioners because of minor psychiatric illness (Cherry & Waldron, 1984) and deaths from presenile dementia (O'Flynn *et al.*, 1987). [The Working Group noted that the confidence intervals were wide and that the results of the latter studies could thus be considered non-positive rather than negative.] Painters were overrepresented among cases of psychomotor epilepsy (Littorin *et al.*, 1988).

[The Working Group noted that reasons for the variable outcome include differences in exposures, i.e., identity of chemicals, intensity and duration. Also, selection bias may have occurred; and the examination methods varied, some possibly being influenced by recent rather than chronic exposures. Finally, the control groups used may have not been appropriate, so that the effects of confounders cannot be ruled out.]

(iv) *Kidneys*

At the beginning of the century, it was claimed that exposure of painters to turpentine caused glomerulonephritis; this association was not firmly established (Chapman, 1941), although the suspicion that a toxic effect of solvents caused clinical disease of the glomeruli remained. Goodpasture's syndrome has been associated with exposure to paint solvents (Klavis & Drommer, 1970; Beirne & Brennan, 1972).

Case studies of glomerulonephritis indicated a possible association with exposure to various solvents, including those in paints (Zimmerman *et al.*, 1975; Ehrenreich *et al.*, 1977; Lagrue *et al.*, 1977; Ravnkov *et al.*, 1979; Finn *et al.*, 1980), although one study that showed a relative risk (RR) of 1.1 (95% confidence interval (CI), 0.4-3.1) did not (van der Laan, 1980).

Most studies on kidney disease in the painting trade have concentrated on solvents. Several solvents are nephrotoxic (Lauwerys *et al.*, 1985). In a study of industrial spray painters exposed to toluene-containing paints, no indication of kidney disease was observed (Greenburg *et al.*, 1942). Later cross-sectional studies using more sophisticated methods revealed only minor effects. Among paint industry workers who were exposed to toluene and xylene, slight haematuria and albuminuria were observed but no effect on concentrating ability or glomerular filtration rate (Askergren, 1981; Askergren *et al.*, 1981a,b,c). These results were interpreted as being a minor effect on the glomeruli. In another study of painters exposed to toluene and xylene, indications of very slight tubular effects were reported (Franchini *et al.*, 1983). In a third study of car painters exposed to low levels of white spirits and

toluene, no such effect was observed (Lauwerys *et al.*, 1985); however, a minor increase in urinary albumin excretion was reported among house painters using mainly water-based paints (Askergren *et al.*, 1988).

Kidney disease may be caused by exposure to lead in paints (Skerfving, 1987; see also IARC, 1980a).

(v) *Liver and gastrointestinal tract*

Slight effects on serum liver enzymes were noted in early studies of groups of industrial spray painters (Greenburg *et al.*, 1942). In other studies of paint industry workers (Lundberg & Håkansson, 1985), of car painters (Kurppa & Husman, 1982), of house painters (Hane *et al.*, 1977) and of subjects with suspected organic solvent poisoning (e.g., car painters; Milling Pedersen & Melchior Rasmussen, 1982), no consistent change in levels of serum liver enzymes was observed.

Lead may cause colic ('painter's colic'), and solvents and arsenic have also been claimed to cause gastrointestinal symptoms among painters and varnishers (Rosen, 1953).

(vi) *Blood and haematopoietic system*

Results obtained from haematological studies of workers in the painting trade vary. In painters who used gasoline as a solvent, a reduction in blood haemoglobin level was observed. Typical levels of aromatic hydrocarbons (one-fifth to one-tenth of the total hydrocarbon content) were 300–800 ppm (Sterner, 1941). Similarly, in later studies of car spray painters exposed mainly to xylene at rather low levels (Angerer & Wulf, 1985) and of house painters who had been exposed to various solvents (Hane *et al.*, 1977), slight decreases in haemoglobin levels were reported. In contrast, in one study of car and industrial spray painters, increased levels of haemoglobin were reported (Elofsson *et al.*, 1980).

In early studies of car spray painters, a slight decrease in white cell counts was observed, with relative lymphocytosis (Lind, 1939). This was probably due to a myelotoxic effect of benzene which was a contaminant of toluene and xylene before 1950–60. However, in one later study of patients with suspected solvent poisoning (mostly house, industrial and car painters), a slight decrease in white cell counts was reported (Milling Pedersen & Melchior Rasmussen, 1982), and in a study of car spray painters, lymphocytosis was observed (Angerer & Wulf, 1985). In contrast, another study of house painters showed no change in white cell counts (Elofsson *et al.*, 1980).

In more recent studies of paint industry workers (Beving *et al.*, 1984b; Lam *et al.*, 1985; Ørbaek *et al.*, 1985) and car painters (Beving *et al.*, 1983), slight decreases in thrombocyte counts were observed; in paint industry workers, the fatty acid composition of platelet membrane was altered (Beving *et al.*, 1988). In a further study of patients with chronic poisoning suspected to be induced by solvents (e.g., car painting), no change in thrombocyte counts was reported (Milling Pedersen & Melchior Rasmussen, 1982).

In painters, lead affects the formation of haemoglobin and red cells in bone marrow and causes haemolysis in peripheral blood (Skerfving, 1987).

(vii) *Other organs*

Some indication has been found that solvents affect muscles (raised serum creatine kinase levels), as seen during short-term exposure of volunteers to white spirits (Milling Pedersen & Cohr, 1984), in workers (e.g., house painters; Milling Pedersen & Melchior Rasmussen, 1982) and in patients with poisoning suspected to be due to solvents (mostly house, industrial and car painters; Milling Pedersen *et al.*, 1980). In the latter study, an increase in the activity of lactic dehydrogenase was observed in muscle biopsy specimens.

Case histories have been reported of subjects who suffered myocardial infarction after exposure to dichloromethane in paint removers (Stewart & Hake, 1976). However, cohort studies of paint industry workers have not indicated an increased risk for cardiovascular disease (Morgan *et al.*, 1981, 1985; Lundberg, 1986).

(viii) *Mortality from conditions other than cancer*

Many of the papers mentioned below are discussed in greater detail in section 3.3. Only statistically significant results are given here.

In one study of US paint industry workers, no increase in the total deaths from diseases of the nervous system was observed over that expected (Morgan *et al.*, 1981), and in a further study a significant decrease was observed (Matanoski *et al.*, 1986). In a study of Swedish painters, there was increased mortality from suicide (Engholm & Englund, 1982; Engholm *et al.*, 1987).

A cohort of Swedish painters showed an increase in mortality from chronic obstructive respiratory disease (Engholm & Englund, 1982), but no such increase was seen in studies of US painters (Matanoski *et al.*, 1986), of US automobile painters (Chiazze *et al.*, 1980) or of US aeroplane spray painters (Dalager *et al.*, 1980) or in two studies of workers in the paint industry (Morgan *et al.*, 1981; Lundberg, 1986).

In two further studies of paint industry workers, no increase in the total number of deaths from diseases of the genitourinary system was observed (Morgan *et al.*, 1981; Lundberg, 1986), although in one of the studies three deaths from infectious urinary tract disease were observed among cleaners in paint manufacture who had been heavily exposed to solvents, while only 0.2 were expected (Lundberg, 1986).

In two studies of paint industry workers, no increase in the total number of deaths from diseases of the gastrointestinal tract was observed (Morgan *et al.*, 1981; Lundberg, 1986). In another study of US painters, a significant decrease in the number of deaths from gastrointestinal disease was observed (Matanoski *et al.*, 1986). Increased mortality from diseases of the oesophagus and stomach has been reported in painters (Engholm & Englund, 1982). There was an indication of an increased rate of liver cirrhosis in one study (Lundberg, 1986), and, in automobile (Chiazze *et al.*, 1980) and aeroplane (Dalager *et al.*, 1980) spray painters, proportionate mortality from liver cirrhosis also appeared to be increased. Similar findings were reported in Swedish house painters (Engholm *et al.*, 1987). Danish house painters did not display an increase in the incidence of cirrhosis (Mikkelsen, 1980). [The Working Group noted that, in interpreting effects on the liver and gastrointestinal tract, the possibility that workers in the painting trade have a higher alcohol consumption than the general population must be considered.]

In one study of paint industry workers, no increase in the total number of deaths from diseases of the blood or blood-forming organs was found (Morgan *et al.*, 1981).

Studies of paint industry workers have not indicated an increased risk for cardiovascular disease (Chiazze *et al.*, 1980; Morgan *et al.*, 1981; Engholm & Englund, 1982; Morgan *et al.*, 1985; Lundberg, 1986; Matanoski *et al.*, 1986). In one study of Danish painters (Mikkelsen, 1980), deaths from diseases of the circulatory system were increased 30% as compared to the general population, but not as compared to a control group of bricklayers. Spray painters in automobile factories showed increased proportionate mortality from hypertensive heart disease (Chiazze *et al.*, 1980).

An increased number of deaths from cerebrovascular disease was observed in paint factory workers (Morgan *et al.*, 1981, 1985). Data on cerebrovascular mortality among aeroplane painters are in accordance with these results but are not significant (Dalager *et al.*, 1980). In a study of US painters, a significant decrease in the number of deaths from cerebrovascular disease was observed (Matanoski *et al.*, 1986).

[The Working Group noted that cohorts of workers in the painting trades may be subject to selection, which may bias the results of mortality studies. Also, in mortality studies, the occupational and disease categories used are broad, decreasing the specificity of the observations.]

(b) *Effects on fertility and on pregnancy outcome*

(i) *Fertility*

McDowall (1985) analysed a 10% sample of 601 526 births within marriage registered in England and Wales in 1980–82 for which the occupation of the father was recorded on the birth certificate. The standardized fertility ratio for men in each of 350 occupational units, defined by the Office of Population Censuses and Surveys, was calculated, taking the value for all occupational groups combined to be 100. Table 21 summarizes the findings in the five occupational groups in which paternal exposure to paint is likely: artists and commercial artists; coach painters; other spray painters; painters and decorators not elsewhere classified, and french polishers; and painters, assemblers and related occupations. Men with occupations classified as 'other spray painters' and 'painters and decorators, and french polishers' had significantly more children than expected on the basis of national rates (standardized fertility ratios, 129 and 141, respectively, based on 694 and 2871 births).

Rachootin and Olsen (1983) carried out a case-control study of 1069 infertile couples and 4305 fertile control couples attending Odense University Hospital, Denmark, in 1977–80. The RRs associated with occupational exposure to 'lacquer, paint or glue' were 1.2 (95% CI, 0.9–1.7) for men with sperm abnormalities, 1.1 (0.7–1.7) for women with hormonal disturbances, 1.4 (0.8–2.6) for women with idiopathic infertility and 1.1 (0.7–1.8) for men with idiopathic infertility.

Bjerrehuus and Detlefsen (1986) reported on a postal survey of 3251 male painters in Copenhagen, Denmark, and 1397 construction labourers. Approximately half responded, and 18% of the painters reported failure to conceive after two years of trials, compared with 10% of the construction workers. Telephone interview with a sample of the painters who had not responded to the postal questionnaire yielded a similar infertility rate.

Table 21. Standardized fertility ratios, sex ratio, percentage of births with birthweights of less than 2500 g, stillbirths, perinatal mortality and infant mortality, according to father's occupation, in occupational units in which exposure to paint is likely; England and Wales, 1981-82^a

Occupational title (Office of Population Census and Surveys, 1970)	No. of births ^b	Standardized fertility ratio	Sex ratio (M:F births)	Births with birthweight < 2500 g (%)	Stillbirths		Perinatal mortality		Infant mortality	
					SMR	No.	SMR	No.	SMR	No.
Artists; commercial artists	373	105	0.884	4.8	75	19	65	20	72	19
Coach painters	20	89	1.000	10.0	73	1	56	1	119	2
Other spray painters	694	129*	1.224*	8.1	112	52	120	71	111	62
Painters and decorators not elsewhere specified; french polishers	2871	141*	1.049	6.6	99	191	99	242	98	224
Painters, assemblers and related occupations	341	100	1.018	7.9	131	30	131	38	144*	38
All occupations	601 526	100	1.061	6.6	100		100		100	

^aFrom McDowall (1985); SMR, standardized mortality ratio

^b10% sample, except for 'all occupations'

*Differs significantly from all occupations ($p < 0.05$)

(ii) *Perinatal toxicity*

Olsen and Rachootin (1983) reported in a letter to the Editor data on 2259 couples who had had a healthy child in 1978–79 at the Odense University Hospital, Denmark. Occupational exposure to various substances was assessed prior to delivery. Exposure to 'lacquer, paint or glue' was reported by 217 mothers and 1512 of their spouses. For maternal exposures, mean birth weights were 64 g less than the average; the authors reported that, after adjustment for maternal age, smoking and drinking habits and time to conception, birth weights were 51 g less than the average ($p = 0.12$). For paternal exposures, the adjusted birth weight of the babies was 14 g above average ($p = 0.56$).

Heidam (1984a,b) carried out a postal survey of the reproductive history of women living in Funen county, Denmark. Female painters were recruited from the local divisions of the trade union, and 76 of 81 (94%) to whom questionnaires were sent replied; among these, 38 pregnancies were reported (0.5 per woman), of which five (13%) were reported to be spontaneous abortions. A 91% response rate was obtained from a reference group of 1571 employed women; among these, 843 pregnancies were reported (0.5 per woman), of which 84 (10%) were reported to be spontaneous abortions. The corresponding RR was 2.9 (95% CI, 1.0–8.8) for painters after controlling for gravidity, pregnancy order and age. When, however, the analysis was done using separate data on births and spontaneous abortions registered in hospital, the corresponding RR was 1.1 (0.4–2.9). The authors suggested that there may have been some reporting bias among the painters.

McDowall (1985) also presented data relevant to perinatal toxicity, including the sex ratio of offspring, percentage of low birth weights and standardized mortality ratios (SMRs) for stillbirths and perinatal and infant deaths, for births in England and Wales in 1980–82 (Table 21). 'Other spray painters' had an elevated ratio of male:female births; but in each of the four other occupational units with exposure to paint, the sex ratio was below average. The infant mortality rate for the offspring of 'painters, assemblers and related occupations' was higher than that of all occupations. Workers in this group would generally be classified in social class IV, in which the infant mortality rate is 115.

Daniell and Vaughan (1988) used records of live births in Washington State, USA, from 1980–83 to compare the outcome of pregnancy in various occupational groups. Among the 1299 live births for which the occupation of father of the child was described as 'painter', the sex distribution and Apgar score at 1 min and 5 min were similar to that found in the 2529 live births for whom the occupation of the father was described as 'electrician' and in 1469 'general controls'. The RR for low birth weight (< 2500 g) among the offspring of painters was 1.1 (95% CI, 0.7–1.5) compared to 'electricians' and 1.4 (0.9–2.1) when compared to 'general controls'.

(iii) *Malformations*

McDowall (1985) also reported on malformations in England and Wales in 1980–82, according to maternal and paternal occupation (Table 22). Overall, there was no excess of malformations, except in the offspring of men in occupations classified as 'painters, assemblers and related occupations'. When specific malformations were considered, there was an excess of polydactyly in the children of men and women with occupations classified as 'paint-

Table 22. Standardized malformation ratios for specified malformations according to occupation, England and Wales, 1980–82^b

Malformation	Artists; commercial artists				Coach painters				Other spray painters				Painters and decorators not elsewhere specified; french polishers				Painters, assemblers and related occupations			
	Father		Mother		Father		Mother		Father		Mother		Father		Mother		Father		Mother	
	Ratio	No.	Ratio	No.	Ratio	No.	Ratio	No.	Ratio	No.	Ratio	No.	Ratio	No.	Ratio	No.	Ratio	No.	Ratio	No.
All malformations	91	46	100	14	102	3	-	0	97	97	100	4	89*	369	100	4	241*	117	100	140
Anencephalus	381	3	-	0	-	0	-	0	120	2	-	0	58	4	-	0	126	1	187	5
Spina bifida	-	0	-	0	-	0	-	0	64	3	-	0	99	19	495	1	314*	7	177	12
Spina bifida and/or anencephalus	102	3	-	0	-	0	-	0	82	5	-	0	84	2	372	1	275*	8	187*	17
Cleft palate and/or cleft lip	93	3	-	0	-	0	-	0	111	7	-	0	77	20	-	0	163	5	111	10
Hiatus hernia and/or diaphragmatic hernia	-	0	-	0	-	0	-	0	-	10	-	0	101	3	-	0	-	0	94	1
Tracheo-oesophageal fistula, oesophageal atresia and stenosis	-	0	-	0	-	0	-	0	138	1	-	0	99	3	-	0	-	0	92	1
Rectal and anal atresia and stenosis	-	0	559	1	-	0	-	0	80	1	-	0	97	5	-	0	166	1	55	1
Malformations of the heart and circulatory system	156	5	-	0	550	1	-	0	80	5	-	0	79	20	-	0	166	5	58	5
Hypospadias, epispadias	54	2	288	3	-	0	-	0	119	9	-	0	78	24	-	0	194	7	83	9
Polydactyly	49	1	-	0	-	0	-	0	164	7	-	0	176*	31	1081*	2	392	8	144	9
Syndactyly	105	2	-	0	-	0	-	0	57	2	-	0	150	22	763	1	459*	8	213*	10
Reduction deformities	-	0	-	0	-	0	-	0	47	1	-	0	82	7	-	0	399*	4	138	4
Exomphalos, omphalocele	-	0	692	1	-	0	-	0	-	0	-	0	39	2	-	0	357	2	223	4
Down's syndrome	90	2	170	1	-	0	-	0	35	1	505	1	60	8	-	0	123	2	72	3

^aFor fathers, standardized malformation ratios are calculated, taking all occupations as 100. For mothers, standardized *proportionate* mortality ratios are calculated, taking all malformations in each occupational group as 100.

^bFrom McDowall (1985)

*Differs significantly from all occupations ($p < 0.05$)

ers and decorators not elsewhere specified, and french polishers' and in children of men whose occupations were described as 'painters, assemblers and related occupations'. Syndactyly was in excess in the offspring of men and women whose occupation was 'painters, assemblers and related occupations'. Reduction deformities were also in excess for paternal exposure but not for maternal exposure. Spina bifida and/or anencephalus were in excess in the offspring of men and women described as 'painters, assemblers, and related occupations'.

Olsen (1983) reported data from the Register for Congenital Malformations in the county of Funen, Denmark, and took details of parental occupation from birth certificates. The authors reported a relative prevalence ratio of 4.9 (95% CI, 1.4-17.1) for congenital malformations of the central nervous system in the group in which the children's fathers were entered as painters in comparison with all other occupations; the ratio for mothers in this category was 0.

(c) *Genetic and related effects*

Haglund *et al.* (1980) studied chromosomal aberrations and sister chromatid exchanges in the lymphocytes of 17 male paint industry workers (exposed to organic solvents) who were presumed to have the highest exposure among a group of 47 paint industry workers employed in seven different factories in southern Sweden. For each exposed person, a control was chosen, matched by sex, age, place of residence (rural/urban) and smoking habits. Most of the controls were also factory workers (storeroom personnel, paint grinders, electricians, drivers, carpenters), but presumably unexposed. For analysis of both chromosomal aberrations and sister chromatid exchange, lymphocytes were cultured for 72 h; 20-25 metaphases were studied for sister chromatid exchange (17 subjects) and 100 for chromosomal aberrations (five subjects with the highest combined exposure). No difference was seen in either parameter; a significant difference in the frequency of sister chromatid exchange was observed between smokers and nonsmokers (0.202 and 0.175, respectively; $p = 0.02$). [The Working Group noted the small number of workers studied for chromosomal aberrations.]

Sister chromatid exchange was studied in the peripheral lymphocytes of 106 members of the International Brotherhood of Painters and Allied Tradesmen in two major US cities (Kelsey *et al.*, 1988). Intensity and duration of chronic exposure to solvents were estimated from interviewer-administered questionnaire data. Eight men reported no occupational history of solvent exposure; 13 allied tradesmen (including dry-wall tapers and paperhangers) reported minimal, indirect exposure to solvents and had no history of direct application of solvent-based materials. Cumulative exposure (CEI) to solvents was estimated for the working lifetimes of 85 painters. Fifty cells from each of 91 individuals were scored for sister chromatid exchange; for the remaining 15 persons, a mean of 21.2 cells per individual was examined. Cultures were incubated for 72 h. There was no elevation in the frequency of sister chromatid exchange attributable to cumulative duration of exposure to solvents or to intensity of exposure over the year prior to blood sampling. Smoking was associated with a significant elevation in the level of sister chromatid exchange (6.75 versus 5.73 in nonsmokers).

3.3 Epidemiological studies of carcinogenicity in humans

(a) *Occupational mortality and morbidity statistics*

Detailed data from some of the studies described below are given in Table 23.

(i) *National studies*

The occupations recorded on a 10% sample of death certificates in England and Wales were used to calculate SMRs for deaths occurring around the time of the 1951 Census (Office of Population Censuses and Surveys, 1958), of the 1961 Census (Adelstein, 1972; Office of Population Censuses and Surveys, 1972), of the 1971 Census (Office of Population Censuses and Surveys, 1979) and of the 1981 Census (Office of Population Censuses and Surveys, 1986). The SMRs for various cancer sites among painters and decorators are listed in Table 23. SMRs for all cancers were consistently above the average and those for lung cancer consistently 40% above the national average: 149 (909 deaths) in 1949–53, 143 (1502 deaths) in 1959–63, 139 (847 deaths) in 1970–72 and 142 (803 deaths) in 1979–80, 1982–83. The proportion of current smokers among painters and decorators was reported to be slightly higher than that in the total population (smoking ratio, 110, based on a sample of 7566 men, including 153 painters and decorators; Office of Population Censuses and Surveys, 1979).

Guralnick (1963) divided specific causes of death in the USA in 1950 by occupation and industry as reported on death certificates and compared them with the expected causes of deaths of all working men as reported in the census from the same year. There were 6145 deaths among white male painters and plasterers in the age group 20–64, with a SMR of 114; selected SMRs are: all cancers, 126 (1016 cases); buccal cavity and pharynx, 137 (41); oesophagus, 109 (25); stomach, 127 (130); lung, 155 (248); kidney, 120 (24); bladder, 146 (38); brain, 134 (39); and leukaemia, 117 (41) (see Table 23). Proportionate mortality ratios (PMRs) were used to test for significance; only that for lung cancer was significant.

Dunn and Weir (1965) established in 1954 a fixed cohort of 68 153 working men engaged in occupations suspected of engendering a risk for lung cancer and followed them for mortality through to 1962 for this report. In this group, 12 572 men were painters and decorators. Information on smoking and occupation was gathered for this population until 1957. The number of deaths in the eight-year follow-up was compared with that among men in California, USA, 1959–61. Painters and decorators had an SMR of 129 (91 observed) for lung cancer; adjustment for smoking resulted in a decrease in the SMR to 114. The SMR for all other cancers was 94 (153 observed). [The Working Group noted that, since other cancers were treated as a group, it is impossible to determine the risk for those at specific sites.]

Howe and Lindsay (1983) followed a cohort comprising 415 201 Canadian men with known occupational histories in 1965–69, which represented 10% of the Canadian labour force. Cancer mortality in this cohort was monitored by record linkage with a Canadian mortality data base containing all deaths registered in Canada for the years 1965–73. The only significantly elevated SMR (285; based on five observed cases) was found for cancer of the buccal cavity and pharynx, except lip, in the occupational group of construction and maintenance painters, paperhangers and glaziers as compared to the mortality of the entire cohort.

Table 23. Cancer mortality or incidence in studies of national statistics and of large occupational cohorts of painting trades

Type of neoplasm	National statistics										Occupational cohorts					
	UK, 1949-53 (OPCS, 1958); 'other', painters and decorators; males, 20-64		UK, 1959-63 (OPCS, 1972); painters and dec- orators; males, 15-64		UK, 1970-72 (OPCS, 1979); painters and dec- orators; males, 15-64		UK, 1979-80, 1972-83 (OPCS, 1986); painters, decorators and french polishers; males, 20-64		USA, 1950 (Gu- ralnick, 1963); painters and plas- terers; white males, 20-64		Sweden, 1958- 71 (Englund, 1980; Engholm & Englund, 1982); painters' union		Denmark, 1970-79 (Olsen & Jensen, 1987); painters (construction)		USA, 1975-79 (Matanoski <i>et al.</i> , 1986); mixed painters	
	SMR ^a	No.	SMR ^a	No.	SMR ^a	No.	SMR ^a	No.	SMR ^b	No.	SIR ^c	No.	SPIR ^d	No.	SMR ^b	No.
All malignant neoplasms	124	2092**	122	2361**	123	1382**	124	1781**	126	1016**	109	647*	NA	110	927**	
Buccal and pharynx	114	16 ^e	78	12	138	10	145	40*	137	41	NA	61	5	NA		
Oesophagus	84	31	115	53	130	47	106	57	109	25	215	17	148	4	NA	
Stomach	122	360**	120	383**	118	174*	113	132	127	130*	[195 24]**	94	11	136	50*	
Colon	106	120	101	123	98	78	88	74	112	77	NA	NA		111	93	
Rectum	107	103	103	100	101	57	128	82	112	47	NA	NA		NA		
Liver and gall-bladder	65	11	100	26	103	9 ^f	NA	NA	NA	NA	200	12 ^g	120	3 ^f	20 ^f	
Larynx	91	21	58	14	127	16	141	21	200	28**	177	14	NA	NA		
Nasal cavity	40	2	120	6	54	2	172	5	NA	NA	NA	125	1	NA		
Lung	149	909**	143	1502**	139	847**	142	803**	155	248**	128	81*	149	79**	118	326**
Prostate	105	39	102	43	97	27	109	38	82	28	[127 124]**	48	8	98	84	
Kidney	86	25	95	39	104	27	NA	48	120	24	NA	61	4	141	27	
Bladder	109	58	118	79	152	66**	116	48	146	38*	NA	112	24	126	40	
Non-Hodgkin's lympho- ma	109	23	95	38	101	26	NA	NA	NA	NA	NA	56	3	NA		
Hodgkin's disease	113	35	90	37	52	10	144	21	129	22	NA	106	3	NA		
Multiple myeloma	NA		106	19	82	11	NA	NA	NA	NA	NA	NA		NA		
Leukaemia	111	50	98	65	125	43	81	33	117	41	173	13 ⁱ	75	5	116	37

^aSMR, standardized mortality ratio; expected numbers based on national rates for working men

^bExpected numbers based on national rates for white males

^cSIR, standardized incidence ratio; expected numbers based on national rates; in square brackets, SMR

^dSPIR, standardized proportional incidence ratio; expected numbers based on the proportions of cancers in all persons registered in the Danish Pension Fund

^ePharynx

^fLiver

^gIntrahepatic bile ducts

^hGall-bladder

ⁱLymphatic leukaemia

*Significant at the $p < 0.05$ level

**Significant at the $p < 0.01$ level

NA, not available; OPCS, Office of Population Censuses and Surveys

In cohort studies and national statistics, information on smoking habits is not usually available. A review addressing the effect of smoking as a confounding variable in studies of occupational groups (Simonato *et al.*, 1988) indicates that smoking has a limited effect on the association between lung cancer and occupational exposures: the estimates might be increased by 20–25%. The estimates in the studies described above were usually increased to a greater extent.

(ii) *Other studies*

As part of the US Third National Cancer Survey, both occupation and industry were identified for each subject based on main lifetime employment, recent employment and other jobs held (Williams *et al.*, 1977). Interviews were obtained for a total of 7518 men and women, representing a 57% response rate. The RRs for cancers at particular sites were estimated for specific occupations. Painting (which included painters, construction workers, paper-hangers, and pattern and model makers) was the main lifetime occupation for 27 men and was associated with an excess RR for lung cancer: 4.2, based on 12 cases ($p < 0.01$). There were two cases of leukaemia (RR, 4.0).

In a study carried out in California, USA, using the death certificates of about 200 000 white men during the period 1959–61 (Petersen & Milham, 1980), the cause of death and usual occupation as reported on the death certificate were used to calculate proportionate mortality ratios (PMRs), standardized for age and year of death. The total number of deaths from all causes among painters was 3558. An elevated PMR for lung cancer was reported among painters, but figures were not available to the Working Group.

In a similar study, Milham (1983) analysed the death records of 429 926 men and 25 066 women during the period 1950–79 in Washington State, USA. The PMR for cancers of the lung, bronchus and trachea was significantly elevated among painters (mainly construction and maintenance painters): PMR, 121, $p < 0.01$ for all ages, 251 observed; PMR, 112, not significant for ages 20–64, 103 observed; and among auto painters and body/fender repairmen: PMR, 184, $p < 0.01$ for ages 20–64, 29 observed; PMR, 148, $p < 0.05$ for all ages, 39 observed. The PMR for gastric cancer was also elevated among painters and body/fender repairmen. Among paperhangers and decorators (painters), the PMR for lung cancer was elevated (139; 21 observed), but not significantly for ages 20–64; it was significant for all ages (PMR, 140; $p < 0.05$; 50 observed). In persons with this occupation, cancers of the bladder and other urinary organs (PMR, 179, $p < 0.05$ for all ages, 11 observed; PMR, 186, not significant for ages 20–64, two observed) and reticulosarcoma occurred in excess.

Dubrow and Wegman (1984) examined cancer mortality patterns by occupation for white males over 20 years old in Massachusetts for 1971–73. Using age-standardized mortality ratios, 397 occupational categories defined from information on death certificates were assessed for their association with increased risk for 62 malignancies. Increased risks (at $p < 0.05$) were apparent for stomach cancer (23 deaths; SMR, 158) in construction and maintenance painters; for cancer of the trachea, bronchus and lung in grouped painters (110 deaths; SMR, 131) and in shipyard painters (nine deaths; SMR, 261); and for laryngeal cancer (ten deaths; SMR, 205), skin neoplasms except malignant melanoma (four deaths; SMR, 492) and prostatic cancer (36 deaths; SMR, 146) in grouped painters. Grouped painters aged 55–74

years had a statistically significant increase in risk for buccal cavity and pharynx (14 deaths; SMR, 222); a nonsignificant excess of lymphomas was seen for men in the age group 20–64 years (eight deaths; SMR, 192).

Pearce and Howard (1986) compared cancer deaths among males aged 15–64 years in New Zealand in 1974–78, for whom occupation had been listed on the death certificate, with a 10% sample of census data. The RR for leukaemia was 2.3 in association with the occupation of painting (eight cases; 95% CI, 1.0–4.6). When adjusted for social class, the RR fell to 2.0 (95% CI, 0.86–3.9).

(b) *Cohort studies or studies within a cohort*

(i) *Painters*

Chiazze *et al.* (1980) studied workers in ten automobile assembly plants in five large companies in the USA. The plants were selected because of large numbers of employees, similar spray-painting operations, geographic dispersion and adequate records. The study was based on 4760 deaths among active and retired workers from 1970 or 1972 through 1976. A total of 4215 decedents were eligible for study, and employee work records were reviewed; for 253, work histories could not be obtained. The analysis was restricted to white males, who comprised about 80% of the decedents; 226 were spray painters. There was no significant excess proportion of deaths from any cause among spray painters, using either external local deaths or internal non-spray painters deaths. Lung cancer (21 deaths), which was the focus of the study, occurred more frequently among spray painters (PMR, 141) than in the local populations but not more frequently than among other automobile assembly workers (PMR, 108). PMRs greater than unity were noted also for leukaemias and lymphomas and for tumours of the brain, prostate, buccal cavity and pharynx. A nested case-control study covered 263 automotive workers who had died from lung cancer; they were matched by age within two years and by plant of employment with 1001 controls who had died of either cardiovascular disease or accidents. Spray painting was associated with a nonsignificant RR of 1.4 for lung cancer, and there was no indication of a dose-response relationship in association with exposure. The RR for those who had first been exposed at least 15 years prior to death was 1.0. The authors noted that individuals who had worked for only a few years may not have been included among the deaths if they had not been identified by an insurance claim in the company beneficiary file.

Englund (1980) and Engholm and Englund (1982) studied a cohort of 30 580 members of the Swedish painters' union from 1966 to 1974 for mortality and to 1971 for cancer morbidity by matching with national registers. The loss to follow-up was 1%. The SMR for all causes among painters was 102 (2740 cases), and the SIR for cancer was 109 ($p = 0.01$; 647 cases). Excesses were seen for cancers of the oesophagus (17 cases; SIR, 215 [95% CI, 124–340]), liver and bile ducts (12 cases; SIR, 200 [103–349]), lung (81 cases; SIR, 128 [106–152]) and larynx (14 cases; 177 [97–297]) and for lymphatic leukaemia (13 cases; 173 [92–296]). In a study based on population-based registries, about 38 000 painters in the 1960 census were linked to the national cancer registry, 1960–73. Among the 2064 cancers in painters, excesses were seen for cancers of the oesophagus (38 cases; SIR, 148) and of the intrahepatic bile duct (eight cases; SIR, 172). There was also a two-fold excess of pleural

tumours based on six cases. The SIR for all cancers was shown to increase with increasing number of years since entry into the union (Engholm & Englund, 1982). The authors suggested in an abstract that smoking habits were no different among painters than among other groups (Engholm *et al.*, 1987).

Dalager *et al.* (1980) examined the risks for cancer among spray painters employed in the aircraft maintenance industry, where there was exposure to zinc chromate primers. Deaths among painters were compared with those expected among US white males using PMRs. The PMRs for all cancers (136, 50 cases) and for lung cancer (184, 21 cases) were significantly raised. The PMRs for cancers at several other sites were increased but not significantly so. The PMR for respiratory cancers increased with duration of employment.

In a study of 2609 male painters belonging to two painters' unions in the Copenhagen area, Mikkelsen (1980) found no increased risk for all cancers combined when the number of cases (82) was compared with those among men in a bricklayers' union (RR, 1.1; 95% CI, 0.8–1.6) or with those among all Copenhagen men (RR, 1.0; 95% CI, 0.8–1.3). Results were not reported for specific sites.

Whorton *et al.* (1983) followed up a group comprising 6424 union members residing in the San Francisco/Oakland Standard Metropolitan Statistical Area, representing six occupations: asbestos workers, bakers, painters, plasterers, plumbers and roofers. Individuals were considered to be members of the cohort if they appeared on union records in July 1976 and 1977. Incident cases of cancer were identified by computer linkage of union rosters to the California Tumor Registry, and the registry's age-, sex- and year-specific incidence rates were used to calculate expected numbers of cancer cases and SIRs. An increased incidence of cancer of the trachea, bronchus, lung and pleura was seen among painters (15 cases; SIR, 199 [95% CI, 112–330]). Relative risks in excess of unity were also observed for leukaemia and for cancers of the prostate and bladder. The authors pointed out that about 15% of all cohort members were of unknown vital status but were assumed to be alive.

In a cohort mortality study of US paint applicators, primarily in new constructions and maintenance, the records of a large international union of painters and allied tradesmen were used (Matanoski *et al.*, 1986). The cohort consisted of 57 175 men who had been born prior to 1940, had had at least one year of union membership, had been members of the union in 1975–79 in four states in different geographical areas, and had died in 1975–79. A total of 1271 (2.2%) individuals were lost to follow-up. Altogether, 5313 deaths occurred (SMR, 88, based on US white male rates). Death certificates were available for all but 288 (5.4%); SMRs were not significantly elevated for cancers at individual sites. Since there was no direct information on individual worker's trades, data from local union chapters were used to define the usual trade of their members; 58% of the cohort belonged to mixed painting locals. Using the US white male population for comparison, significant excess mortality ratios were seen in local chapters for painters for all malignant neoplasms (SMR, 110; 95% CI, 103–117), stomach cancer (136; 101–180) and lung cancer (118; 106–132), and nonsignificant ratios for cancers of the large intestine (111; 90–136), liver (156; 95–241), bladder (126; 90–172) and kidney (141; 93–205) and for leukaemia (116; 82–160) (see also Table 23). When the risks of men in local mixed painting chapters were compared with those of men in special-

ty locals, the mixed painters had significantly higher mortality from all causes, from malignant neoplasms, from lung cancer, from bladder cancer and from leukaemia. [The Working Group noted that the fact that all painters had to have been active dues-paying members at some time during the follow-up period would tend to have enhanced the 'healthy worker' effect in this population.]

A nested case-control study was conducted of lung cancer incidence in the New York unions included in the study described above (Stockwell & Matanoski, 1985). The 124 male lung cancer cases were identified through the New York State Cancer Registry, and 371 controls without cancer were selected randomly from the union membership and stratified by birth date and geographical location of the unions. Responses to questionnaires on work history, work environment and life-style factors were received from 69 (66%) of the cases and 182 (59%) of the controls; of these, 65 (94%) and 55 (33%) were completed by a proxy for cases and controls, respectively. Painting as the reported usual trade was associated with a high risk (RR, 2.8; 95% CI, 1.5-5.2); high risks were also seen for work in allied trades: painter as a union speciality (RR, 3.2; 95% CI, 1.4-7.1) and ever having worked as a painter (RR, 2.6; 95% CI, 1.3-4.9). In the 57 cases for which the information was available, 53 men were reported to have used spackling compounds (probably containing asbestos), compared with 112 of 161 controls (RR for spackling, 5.2; 95% CI, 1.9-14.5). The authors attempted to adjust for several variables, including asbestos exposure (on the basis of use of spackling compounds). The risk for lung cancer among painters who never wore a respirator remained high (5.4; 95% CI, 1.0-29.3). [The Working Group noted that a high proportion of cases reported using spackling compounds and questioned the accuracy of information obtained from a proxy regarding use of painting materials and of respirators.]

All 93 810 incident cases of cancer recorded in 1970-79 at the Danish Cancer Registry were linked with information on longest employment held submitted by the Supplementary Pension Fund (Olsen & Jensen, 1987). The standardized proportionate incidence ratios (SPIRs) for cancer were reported for each cancer site in each industry and occupation on the basis of the expected proportion of that cancer in all industries. Painters in the construction industry had an increased proportion of lung cancers compared to people in other occupations (SPIR, 149; 95% CI, 119-185; based on 79 cases). Workers in the paint, varnish and lacquer manufacturing industries had an increased proportion of cancers of the nasal cavity and sinus, with a SPIR of 620 (95% CI, 155-2480; based on two cases). In a follow-up study of cases registered through 1984 (Olsen, 1988), the SPIR was reduced to 401 (67-1324) based on two cases of sinonasal cancer. Car painters had a SPIR of 1403 (198-9958) for nasal cavity and sinus cancers based on one case. Several other proportions were above one for these three groups, but the excesses were not significant (Olsen & Jensen, 1987).

(ii) *Paint manufacturers*

Bertazzi *et al.* (1981) followed a small cohort of 427 workers employed in paint manufacturing in Italy. The workers had to have been employed for at least six months at any time from 1946 through 1977 to be eligible for inclusion and were followed for 1954-78. The follow-up was 97.7% complete. There was a significant excess of all cancers in this population (18 cases; SMR, 184; 95% CI, 112-285) when national rates were used as the comparison.

Lung cancers occurred at significant excess when either national (eight cases; SMR, 334; 106–434) or local rates (227; 156–633) were used as a standard, and the risk increased with length of exposure and with latency. These workers were exposed to asbestos as well as to chromate pigments.

A similar study of a larger cohort of 16 243 US male workers in the paint and coating manufacturing industry was reported by Morgan *et al.* (1981). These men had been employed for one year or more after January 1946 in 12 large and 20 medium to small companies and were followed through 31 December 1976. Only plants that retained personnel records for at least 15 years were eligible for the study, and out of 47 eligible plants the 32 largest were finally studied. The overall follow-up rate of the cohort was about 94%. Death certificates could not be obtained for 8.2% decedents. There were 2633 deaths in all (SMR, 86). The cohort was divided into seven subgroups on the basis of their exposures as determined from individual job histories; individuals could appear in multiple exposure groups. Deaths from cancers of the colon and rectum occurred at higher rates in the total population than expected on the basis of numbers among US white males (colon: 65 cases; SMR, 138 [95% CI, 107–176]; rectum: 26 cases; SMR, 139 [91–204]). The risk for respiratory cancers, which was a major focus of the study, was not excessive in this population (SMR, 98; 160 cases); information on smoking habits was not available. Deaths from cancer of the liver and biliary passage occurred more frequently than expected in the subgroups of workers potentially exposed to pigments (seven cases; SMR, 273 [108–555]) and lacquer (five cases; SMR, 255 [81–583]). The SMR for leukaemia was 212 (eight cases [92–418]) in the subgroup of workers exposed to lacquer. A further report on this study (Morgan *et al.*, 1985) provided little additional information.

A small cohort of 416 men who had worked for five years or more in the Swedish paint manufacturing industry during the period 1955–75 were followed for mortality in the years 1961–81 (Lundberg, 1986). Reference numbers were taken from national statistics. Subjects were categorized into lower and higher exposure levels according to duration and intensity of exposure. Overall mortality was low (96 cases; SMR, 88), as was mortality from all cancers (22 cases; SMR, 84; 95% CI, 52–127) and from lung cancer (three cases; SMR, 63; 95% CI, 12–184). The SMR for multiple myeloma was 549 (three cases; 95% CI, 113–1606) and that for cancer of lymphatic and haematopoietic tissues 212 (five cases; 95% CI, 68–496). The three cases of multiple myeloma occurred in workers in the higher exposure category.

(c) *Case-control studies*

(i) *Cancers at multiple sites*

Cancer cases recorded at a cancer centre in New York State, USA, in 1956–65 were compared with all patients with non-neoplastic lesions in regard to occupations related to inhalation of combustion products or chemicals and to personal characteristics (Viadana *et al.*, 1976; Decouflé *et al.*, 1977; Houten *et al.*, 1977). The information was obtained through an interview at the time of admission for all patients. Each of the 11 591 white male subjects was included for analysis for each occupation held; specific occupations were compared with those of an unexposed clerical group. Painters were analysed as a subgroup of people with chemical exposures and as a subgroup of those with metal-related occupations. Cancer sites

for which RRs were increased were: lung (42 cases; RR, 1.7; $p = 0.02$), stomach (eight cases; 2.4; $p = 0.05$), oesophagus (seven cases; 3.0; $p = 0.03$), prostate (nine cases; 1.9), bladder (16 cases; 1.6), kidney (four cases; 2.6) and melanoma (two cases; 3.2). The highest RR was seen in the age group below 60 years for stomach cancer (12.6); for oesophageal cancer, the risk was greater for the age group above 60 years (3.8). These two ratios were even higher among painters with five or more years of exposure (16.6 and 6.9, respectively). For lung and prostate cancer, no such dose-response relationship was observed. The elevated lung cancer risk among painters was no longer significant after adjustment for smoking and age (RR, 1.7). The author noted that the risk for stomach cancer was elevated in more than half of the occupations, which might be explained by the eastern European origin of the workers. No adjustment was made for alcohol drinking (see also Tables 24 and 25).

Coggon *et al.* (1986a,b) identified all cases of cancer in three English counties where chemical, metal and vehicle production industries were situated, using hospital and cancer registration records for the period 1975–80. Males aged 18–54 were included in the study. Occupational and smoking histories were obtained either by mailed questionnaires (response rate, 52.1%) or from information in hospital records or on death certificates. A total of 2942 cancer cases were identified, and cases of cancer at 15 specific sites were compared with those at all other sites with regard to occupation. Data were corrected for age, residence, source of history and smoking. Laryngeal cancer was more likely to be associated with painting and decorating (RR, 3.4; 95% CI, 1.3–9.0; six cases) than with other occupations; bronchial cancer was also associated with painting, the RR being 1.3 (20 cases; see also Table 24). A borderline significant association was seen for cancer of the stomach (RR, 2.3; 95% CI, 1.0–5.0); other sites for which the RR was above unity were oral cavity (RR, 1.9; five cases), skin (RR, 1.4; four cases), testes (RR, 1.9; nine cases) and malignant melanoma (RR, 1.6; four cases). The authors commented that five patients with testicular cancer had worked as paint sprayers, which results in a RR of 4.9 (95% CI, 1.3–18.2). A nonsignificant RR of 0.7 was found for bladder cancer (see also Table 25).

In the same area of the UK, Magnani *et al.* (1987) examined occupations associated with cancer at five sites – oesophagus, pancreas, melanoma, kidney and brain. Deaths from these cancers in men aged 18–54 for the period 1959–63 and 1965–79 were matched by year of death, age at death and residence to those among four controls who had died from other causes. Occupation and industry were identified from death certificates. No significant risk for any of the cancers was associated with exposure to painting and decorating; however, the RR for oesophageal cancer was 2.0 (95% CI, 0.8–4.9) and that for brain cancer, 1.4 (95% CI, 0.7–2.8). The investigators also described exposures for each occupation, summed these across occupations, and examined the risks of these substances as they relate to the cancers. In this analysis, paints were associated with only a small increase in RR for three cancers – oesophageal and brain cancers and melanoma; none of the associations is significant. The authors noted that only the most recent full-time job was recorded on the death certificate. No adjustment was made for smoking or alcohol drinking.

Table 24. Case-control and other studies of lung cancer among persons exposed in paint manufacture and painting

Reference	Location, time	Type of controls	Source	Exposure	No. of cases (no. of painters)	RR	95% CI	Comments
<i>Case-control studies</i>								
Wynder & Graham (1951)	USA, NG	[Unclear]	Interview		857 (200 fume-exposed; 11 painters)	NG	NG	
Breslow <i>et al.</i> (1954)	USA, 1949-52	Hospital	Interview	Construction and maintenance painters for ≥ 5 years	518 (22)	1.9	0.93-3.8	Not adjusted
Menck & Henderson (1976)	USA, 1968-70	Estimated population by industry	Death certificates, hospital records	Painter at diagnosis	2161 (45)	SMR, 158	NG	Significant; adjusted for age
Milne <i>et al.</i> (1983)	USA, 1958-62	Deaths from other causes (except pancreas, bladder, nasal, kidney, haematopoietic)	Death certificates, occupation	Painter	925 men (24)	1.8	NG	Significant ($p < 0.01$); adjusted for age
Kjuus <i>et al.</i> (1986)	Norway, 1979-83	Hospital	Interview and worksite records	Paint manufacture	(3)	0.7	NG	Not significant
				Painting and paper-hanging	176 men (5)	1.7	0.4-7.3	Occupation is longest job held; considered exposed if ≥ 3 years; adjusted for smoking
Lerchen <i>et al.</i> (1987)	USA, 1980-82	Population and rosters of elderly	Interview	Paints, glues, lacquer	17	1.2	0.6-2.6	
				Ever construction painters	333 men (9)	2.7	0.8-8.9	Adjusted for age, ethnicity and smoking
				Asbestos	40	1.1	0.7-1.7	

Table 24 (contd)

Reference	Location, time	Type of controls	Source	Exposure	No. of cases (no. of painters)	RR	95% CI	Comments
Siemiatycki <i>et al.</i> (1987a)	Canada, 1979-85	Other cancers	Interview	Listed as white spirits, but in exposed group construction is 21% of total, mostly painters	857 males	1.1	0.8-1.4	Adjusted for age, socio-economic status, ethnicity, cigarette smoking, blue/white collar; 90% CI
					159 (36)	1.2	1.0-1.5	
					359 (92)	1.0	0.7-1.3	
					162 (37)	0.8	0.6-1.1	
					177 (32)	1.7	1.2-2.3	
			Long duration, high exposure	44	1.7	1.2-2.3		
			Construction workers		1.4	NG		
Levin <i>et al.</i> (1988)	China, 1984-85	Population	Interview	Ever painter	733 men (15)	1.4	0.5-3.5	Questionable trend; adjusted for age and smoking
Ronco <i>et al.</i> (1988)	Italy, 1976-80	Deaths without smoking-related diseases	Interview	Painter	164 men (5)	1.3	0.43-4.1	Adjusted for age, smoking and other employment in suspect high-risk occupations
<i>Multisite case-control studies</i>								
Viadana <i>et al.</i> (1976); Decouflé <i>et al.</i> (1977); Houten <i>et al.</i> (1977)	USA, 1956-65	Noncancer admissions	Interview at admission	Painter	(42)	1.7	NG	Significant; adjusted for age; non-significant when adjusted for smoking and age
Coggon <i>et al.</i> (1986a)	UK, 1975-80	Other cancers	Interview	Painter	738 men (20)	1.3	NG	Adjusted for age, smoking, residence, respondent

RR, relative risk; CI, confidence interval; NG, not given; SMR, standardized mortality ratio

Table 25. Case-control studies of lower urinary tract cancer among persons exposed in paint manufacture and painting

Reference	Location, time	Type of controls	Source	Exposure	No. of cases (no. of painters) ^a	RR	95% CI	Comments
Wynder <i>et al.</i> (1963)	USA, 1957-61	Hospital, without smoking-related disease	Interview	Ever painter	300 (18)	[2.2]	[1.0-4.5]	No adjustment for smoking
Cole <i>et al.</i> (1972)	USA, 1967-68	General population	Interview	Painter	461 (28 men)	1.2	0.71-1.9	Adjusted for age and smoking
Howe <i>et al.</i> (1980)	Canada, 1974-76	Neighbourhood	Interview	Commercial painting	480 men (≥ 24)	1.0	0.6-2.3	Unadjusted. After correction for exposure to other suspect 'high-risk' industry, RR for spray painter, 1.0
				Ever spray painting	(≥ 16)	1.8	0.7-4.6	
Silverman <i>et al.</i> (1983)	USA, 1977-78	Population	Interview	Ever painter	303 men (15)	1.0	0.5-2.2	Unadjusted
				Car painter	(3)	0.5	0.1-2.1	
				Paint manufacture	(1)	0.2	0-2.2	
Schoenberg <i>et al.</i> (1984)	USA, 1978-79	Population	Interview	Ever painter	658 men (34)	1.4	0.85-2.3	Adjusted for age, smoking and other employment
				Paint exposure	(111)	1.6	1.2-2.1	
Vineis & Magnani (1985)	Italy, 1978-83	Hospital; other urological and surgical	Interview	Painter in building industry	512 men (12)	1.0	0.40-2.2	Adjusted for age and smoking
				Car painter ≥ 5 years	(7)	2.0	0.60-7.0	
				Carpentry painter	(1)	0.6	0.04-8.4	
				Spray painter in different industries	(2)	1.2	0.20-5.8	
Morrison <i>et al.</i> (1985)	USA, UK, Japan, 1976-78	Population	Interview	Paint and paint manufacture	USA, 430 (35)	1.5	0.9-2.4	Adjusted for age and smoking; 90% CI
					UK, 399 (23)	0.7	0.5-1.2	
					Japan, 226 (5)	0.7	0.3-1.7	
Claude <i>et al.</i> (1988)	FRG, NG	Hospital urological and homes for elderly	Interview	Ever painter	531 men (15)	1.3	0.59-2.7	Trend, $p = 0.04$ for exposure to spray paints
				Lacquer and paint	(78)	1.5	1.1-2.2	
				Spray paints	(52)	2.9	1.7-4.9	
Jensen <i>et al.</i> (1987)	Denmark, 1979-81	Population	Interview	Different painting industries Painter 10 years	371 (13)	2.5 1.4	1.1-5.7 1.0-1.9	Adjusted for age, sex and smoking

Table 25 (contd)

Reference	Location, time	Type of controls	Source	Exposure	No. of cases (no. of painters) ^a	RR	95% CI	Comments
Iscovich <i>et al.</i> (1987)	Argentina, 1983-85	Neighbourhood and hospital	Interview	Ever painter	117 (3)	0.55	[0.12-2.5]	Adjusted for age and tobacco smoke, pooling the two control groups
Schiffers <i>et al.</i> (1987)	Belgium, 1984-85	Population	Interview	Painter in high-risk occupation	74 (NG)	NG	NG	No increased risk reported
Risch <i>et al.</i> (1988)	Canada, 1979-82	Population	Interview	Exposed to paints in full-time job at least 6 months, 8-28 years before diagnosis	781 (204 men, 14 women)	1.1 3.9	0.77-1.6 0.9-26.7	Adjusted for smoking
				Commercial painting	(49 men)	0.90	0.39-2.1	
				Spray painting	(67 men)	0.91	0.48-1.7	
Siemiatycki <i>et al.</i> (1987a)	Canada, 1979-85	Other cancers	Interview	Listed as white spirits, but in exposed group construction is 21% of total, mostly painters	486 (91)	1.0	0.8-1.2	Adjusted for age, socioeconomic status, ethnicity, cigarette smoking, blue/white collar work; 90% CI
<i>Multisite studies</i>								
Coggon <i>et al.</i> (1986b)	UK, 1975-80	Other cancers	Interview	Painter	179 (10)	0.7	NG	Adjusted for age, smoking, residence, respondent; bladder and renal pelvis; men aged 18-54 only
Viadana <i>et al.</i> (1976); Decouflé <i>et al.</i> (1977); Houten <i>et al.</i> (1977)	USA, 1956-65	Noncancer admissions	Interview at admission	Painter	(16)	1.6	NG	Not significant

^aIf only discordant pairs noted, no. of painters \geq number of discordant pairs given

RR, relative risk; CI, confidence interval; NG, not given

[The Working Group noted that the populations studied by Coggon *et al.* (1986a,b) and Magnani *et al.* (1987) may overlap and that only deaths in relatively young men were considered.]

(ii) *Cancer of the lung*

These studies are summarized in Table 24.

In an early descriptive study, Wynder and Graham (1951) studied a total sample of 857 incident cases of lung carcinoma diagnosed in one hospital in St Louis, MO, USA, over an unspecified period. Of 200 who were 'believed or known to have been exposed to irritative dusts and/or fumes', 11 were painters. [The Working Group found it difficult to clarify the information on the comparison groups.]

Breslow *et al.* (1954) identified 518 cases of lung cancer in 11 Californian hospitals during the period 1949–52. Controls were selected from patients admitted to the same hospital for a condition other than cancer or a chest disease, and matched for age, sex and race. Detailed occupational and smoking histories were obtained by interview. The authors reported that 22 cases had been employed as construction or maintenance painters for at least five years, as had 12 controls [RR, 1.9; 95% CI, 0.93–3.8]. Smoking was not controlled for, although smoking histories had been recorded.

Menck and Henderson (1976) identified deaths from lung cancer for the years 1968–70 (2161 cases) and incident cases of lung cancer for the years 1972–73 (1777 cases) from the Los Angeles County Cancer Surveillance Program. Both were classified by occupation and industry on the basis of either death certificates or hospital records. Of the 3938 subjects, 689 had no reported occupation and 1222 no reported industry of employment. Employment of the population aged 20–64 was estimated from a sample of the population in the 1970 census, and the risk of lung cancer for each occupation was compared to the risk in the total population. The SMR for lung cancer in painters was significantly elevated (45 deaths; SMR, 158; $p < 0.01$; see also Table 24).

Milne *et al.* (1983) compared the occupation and industry of 925 (747 male and 178 female) cases of lung cancer in Alameda County, California, USA, 1958–62, with those of people who had died of other cancers. Usual occupation and industry as stated on the death certificate were coded using the US census classification. When occupations were examined separately, male painters had a significantly increased risk for lung cancer (24 cases) when compared either with all cancer deaths (RR, 1.7; $p < 0.05$) or with those dying of cancers other than of the pancreas, nasal sinus, kidney, bladder, bone and haematopoietic organs (RR, 1.8; $p < 0.01$). There was no increased risk associated with employment in the paint manufacturing industry (RR, 0.7; three cases; see also Table 24).

A study of 176 male incident lung cancer cases, under 80 years of age, admitted in 1979–83 to two hospitals in two neighbouring counties in Norway was conducted by Kjuus *et al.* (1986). Controls were matched on age through admission lists or from the same department records; persons with physical or mental handicaps, general poor health or an admission diagnosis of chronic obstructive pulmonary disease were excluded from the control group. Occupational histories were determined by interview and work site records then coded by job title and separated into three groups according to potential exposure to lung

carcinogens, which included painting and paints. Three years was considered to be the minimal exposure classified as positive, and occupation was classified as the longest job held; exposures were included only up to 1970. Within the group, the RR for painting and paperhanging was 1.7 (95% CI, 0.4–7.3; five cases), adjusted for smoking. The RR for lung cancer associated with exposure to paints, glues and lacquer was 1.2 (95% CI, 0.6–2.6; 17 cases), adjusted for smoking, in comparison with all other subjects.

Occupational histories obtained by interview were compared in a case-control study of 506 lung cancer patients (333 men and 173 women) diagnosed in 1980–82, according to the population-based New Mexico Tumor Registry, and 771 controls selected through random telephone numbers or from rosters of elderly (Lerchen *et al.*, 1987). Next-of-kin provided the information for half of cases and 2% of controls. Jobs held by individuals from age 12 years were classified according to an a-priori list of potentially hazardous occupations. Construction workers and painters were included in high-risk occupations; employment for one year or more was classified as ever having been employed in an industry. The RR for lung cancer in men associated with employment as a construction painter was 2.7 (nine cases; 95% CI, 0.8–8.9) compared to never having been employed in that occupation and adjusted for age, ethnicity and smoking.

In the study of Siemiatycki *et al.* (1987a,b), described in detail in the monograph on some petroleum solvents (p. 70), construction workers exposed to white spirits, many of whom were painters, were described as having an excess risk for lung cancer (RR, 1.4 [numbers not given]).

In a cancer registry-based case-control study, Levin *et al.* (1988) identified 833 male lung cancer cases diagnosed between February 1984 and February 1985 in Shanghai, China, and 760 randomly selected male controls from the general urban Shanghai population, frequency matched within five-year age strata. Personal interviews to obtain occupational and smoking histories were obtained for 733 cases and 760 controls. More than 60 industries and occupations were examined; ever *versus* never having worked as a painter was associated with a RR, adjusted for age and smoking, of 1.4 (95% CI, 0.5–3.5). The RR varied according to duration of employment as a painter as follows: < 10 years, 1.9 (seven cases); 10–19 years, 2.8 (two cases); 20–29 years, 2.2 (five cases); ≥ 30 years, 0.3 (one case; questionable trend). The authors cited multiple comparisons and the use of broad occupational groups as limitations of the study.

Ronco *et al.* (1988) reported a population-based case-control study from two areas in northern Italy which included 164 male lung cancer cases identified from death records during 1976–80 and 492 controls who had died of conditions other than chronic lung disease or smoking-related cancers. Information on smoking and occupation was obtained through interviews of next-of-kin. Many exposures suspected of increasing the risk for lung cancer were evaluated, and individuals who had not held any job in any industry that was associated with exposure to a known or suspected lung carcinogen were classified as nonexposed. The RR for painters, adjusted for age, smoking and employment in other studied exposures, was 1.3 (five cases; 95% CI, 0.43–4.1).

Malker *et al.* (1985) examined the risk for pleural mesothelioma in relation to occupational exposures, including painting. The investigators used the Swedish population-based registries to link incident cancer cases during 1961–79 with 1960 census data on occupation and industry. Altogether, 318 cases of pleural mesothelioma occurred. Standardized incidence ratios (SIR) were calculated for occupations and industrial categories. For workers in the construction industry as a whole, a significant SIR of 1.6 was seen based on 63 cases; painting as a specific industry comported a higher significant SIR (2.9, based on 13 cases); painters and paperhangers as a specific craft showed an SIR of 2.0 (based on 12 cases), which was significant. [The Working Group noted that painters in the construction industry are probably exposed to asbestos.]

(iii) *Cancer of the larynx*

A case-control study of incident laryngeal cancer was carried out by Brown *et al.* (1988) in Texas. Cases consisted of all diagnoses of primary laryngeal cancer among white males aged 30–79 selected from 56 participating hospitals, comprising 220 living cases and 83 dead cases identified during the period 1975–80. Controls consisted of an equal number of white males without respiratory cancer selected from various sources and frequency matched on age, vital status, ethnicity and county of residence. Occupational exposures were examined, controlling for cigarette smoking and alcohol consumption. The RR for painters was elevated (11 cases; RR, 2.3; 95% CI, 0.84–6.3), and a significantly elevated risk was found for workers reportedly exposed to paint (32 cases; RR, 1.8; 1.0–3.2). No clear pattern was evident by duration of exposure.

(iv) *Cancer of the urinary tract*

These studies are summarized in Table 25.

Wynder *et al.* (1963) examined occupational and other risk factors associated with bladder cancer in 300 male patients from seven New York hospitals in 1957–61. Controls consisted of an equal number of male hospital patients who did not have myocardial infarction or cancers of the respiratory system or upper alimentary tract and were matched by age and time of admission. Interviews were conducted directly with the patients. The investigators reported 18 painters among cases and 12 among controls. [The Working Group calculated the RR to be 2.2 (95% CI, 1.0–4.5) for the group that had ever worked as a painter; no adjustment was made for cigarette smoking.]

Cole *et al.* (1972) conducted a case-control study of transitional- or squamous-cell carcinoma of the lower urinary tract in eastern Massachusetts using newly diagnosed cases aged 20–89 during an 18-month period ending 30 June 1968 (Cole *et al.*, 1971). Out of 668 cases ascertained, a random sample of 510 was selected for interview; a usable occupational history was obtained for 461. Controls were selected from the general population of the same area and matched on age and sex. Certain occupations (including painting) were classified as 'suspect'; and each of these groups was compared to nonsuspect industries. The RR for lower urinary tract cancer in male painters, adjusted for age and smoking, was 1.2 (28 cases; 95% CI, 0.71–1.9).

Howe *et al.* (1980) conducted a case-control study of bladder cancer in three areas of Canada; they identified 821 cases through provincial cancer registries in 1974–76 and

matched them by age, sex and neighbourhood to 821 controls. Personal interviews were obtained for 632 cases (480 men and 152 women; 77%) and an equal number of controls. Among men, working as a painter was not associated with a risk: the RR for commercial painting was 1.0 (24 cases in discordant pairs; 95% CI, 0.6–2.3); that for spray painting was 1.8 (16 cases in discordant pairs; 0.7–4.6), which was reduced to 1.0 after correction for exposure in other suspect 'high-risk' industries.

As part of the US National Bladder Cancer Study, Silverman *et al.* (1983) conducted a population-based case-control study of bladder cancer in the Detroit, MI, USA, area. They identified 420 male cases diagnosed with transitional- or squamous-cell carcinoma of the lower urinary tract aged 21–84 between 1977–78; interview was obtained for 339 (81%), but the analysis was restricted to 303 white males. Controls were 296 white males stratified for age who were selected from a random digit-dialling survey for those under age 65 and from a random sample of the Health Care Financing Administration lists for those over 65. Employment was measured as 'ever' or 'usual' occupation or industry; 'usually unexposed' were those not employed in the industry of interest. The findings suggest no increased risk for bladder cancer for painters in general (15 cases; RR, 1.0; 95% CI, 0.5–2.2), for painters in the automobile industry (three cases; 0.5; 0.1–2.1) or for paint manufacturers (one case; 0.2; 0–2.2).

A similar case-control study of bladder cancer in 658 white male incident cases aged 21–84 during 1978–79 and of 1258 population controls was conducted in New Jersey, USA, by Schoenberg *et al.* (1984). Controls were selected as by Silverman *et al.* (1983). The RR for bladder cancer in men ever employed as painters, adjusted for age, was 1.4 (34 cases; 95% CI, 0.85–2.3). When occupations were classified by materials used, paint exposure was associated with a risk for bladder cancer (111 cases; RR, 1.6; 95% CI, 1.2–2.1). The risk was higher for those first exposed under age 41 and did not increase with duration of exposure.

A case-control study of bladder cancer in Italy (Vineis & Magnani, 1985) involved 512 male cases aged under 75 between 1978–83 and 596 hospital controls. The controls were matched by age and were subjects with benign urological conditions or surgical conditions. Occupational and smoking histories were obtained by interview. No increased risk was seen for painters in the building industry (RR, 1.0; 95% CI, 0.40–2.2; 12 cases), painters in carpentry (RR, 0.6; 0.04–8.4; one case) or spray painters (RR, 1.2; 0.20–5.8; two cases), but the RR for car painters was 2.0 (95% CI, 0.60–7.0; seven cases).

Morrison *et al.* (1985) examined 15 occupations and the risk for lower urinary tract cancer in Nagoya, Japan (1976–78), Manchester, UK (1976–78), and Boston, USA (1976–77), using incident male cases aged 21–89 and population-based controls. They identified 741 cases in Boston, 577 in Manchester and 348 in Nagoya. Interviews were obtained for 81% of the cases in Boston, 96% in Manchester and 84% in Nagoya; the corresponding figures for the controls were 80%, 90% and 80%. The analysis was limited to 430 cases and 397 controls in Boston, 399 cases and 493 controls in Manchester and 226 cases and 443 controls in Nagoya, for whom smoking histories were known. Occupational exposure to paint or paint manufacture was associated with a risk of bladder cancer only in the Boston population (35 cases; RR, 1.5; 90% CI, 0.9–2.4). This ratio was controlled for age and smoking history. [The

Working Group noted that no specific information was available on how the controls were selected.]

Two publications from the Federal Republic of Germany (Claude *et al.*, 1986, 1988) reported two hospital-based case-control studies of tumours of the lower urinary tract. A total of 340 men and 91 women with such cancer between 1977-82 were matched by age and sex to either hospital patients primarily from urology wards or, for those over 65, to people in homes for the elderly. Subjects were interviewed about occupations, specific exposures and life-style factors. There was no reported excess risk for the occupational category of painting, but the RRs associated with specific exposures suggested a risk of painting in men. Spray painting was associated with an increased risk for cancer of the lower urinary tract (RR, 4.7; 95% CI, 2.1-10.4; 28 cases in discordant pairs), as was exposure to lacquer (RR, 1.6; 95% CI, 0.98-2.5; 45 cases in discordant pairs; Claude *et al.*, 1986). In order to examine occupational risks more extensively, an additional 191 male cases were included, to make a total of 531 (Claude *et al.*, 1988). Painting as an occupation was associated with an increased risk for bladder cancer (RR, 1.3; 95% CI, 0.59-2.7; 15 cases). An examination of the specific exposures indicated significant excess risks for cancer of the lower urinary tract for any exposure to spray paints (RR, 2.9; 95% CI, 1.7-4.9; 52 cases), to lacquer and paints (RR, 1.5; 95% CI, 1.1-2.2; 78 cases) or to chromium/chromate (RR, 2.2; 95% CI, 1.4-3.5). After correction for smoking, a significant trend of increased risk with increasing duration of exposure for individuals exposed to spray paints and chromium/chromate could be seen. [The Working Group questioned the choice of controls and considered that there may have been overlap between the exposure categories.]

Jensen *et al.* (1987) carried out a case-control study of bladder cancer in Denmark and interviewed 371 patients with invasive and noninvasive lesions diagnosed during 1979-81. The occupations of cases were compared with those of 771 controls selected from residents in the same area. Detailed occupational histories were taken, which included industry, type and place of work and duration; the information was coded according to industry. Significantly more cases than controls were employed in furniture lacquering and painting, industrial painting, sign-post painting, painting firms or car painting (13 cases; RR, 2.5; 95% CI, 1.1-5.7). Employment as a painter for ten years gave a RR of 1.4 (95% CI, 1.0-1.9).

Iscovich *et al.* (1987) performed a case-control study of 117 bladder cancer cases diagnosed in Argentina in 1983-85 and individually matched on age and sex to one neighbourhood and one hospital control. Hospital controls were selected from the same hospital as the case; about 12% of patients had diseases known to be associated with tobacco smoking. Neighbourhood controls were selected from among persons living in the same street block as the cases. A detailed questionnaire, containing information on smoking, demographic, socioeconomic and medical variables and occupational history for the three occupations of longest duration as well as the most recent one was administered. No increased risk for bladder cancer was observed among painters (three cases; RR, 0.55; [95% CI, 0.12-2.5]).

A pilot case-control study of bladder cancer in Belgium in 1984-85 (Schiffers *et al.*, 1987) included 74 cases and 203 population controls selected from electoral rolls and matched for age and sex. While cases were interviewed by the investigators, most of the

controls were interviewed by others. A group of 16 jobs, including painting, were defined as hazardous and associated with a high risk for bladder cancer, but exposure to painting as a specific job did not show a significant excess.

A case-control study from Denmark (Jensen *et al.*, 1988) concentrated on cancers of the renal pelvis and ureter. The 96 cases, aged below 80, were identified from 27 hospitals in 1979-82, and three hospital controls were matched to each case on hospital, age and sex. Patients with urinary tract and smoking-related diseases were not eligible as controls. Questionnaire data on smoking and on occupation and occupational exposures were obtained. An elevated risk for upper urinary tract cancer was associated with occupational exposure as painter or paint manufacturer (RR, 1.8, adjusted for sex and lifetime tobacco consumption; 95% CI, 0.7-4.6; ten cases).

A case-control study of bladder cancer was carried out during the period 1979-82 in Alberta and in Toronto, Ontario (Risch *et al.*, 1988). Cases aged 35-79 were identified through a cancer institute, from a province-wide tumour registry in Alberta, and through review of hospital records in Ontario. Interviews were carried out with 835 (67%) of the cases (826 histologically verified) and 792 (53%) of the controls about jobs in 26 industries that had previously been examined in studies of bladder cancer, and on occupational exposures to fumes, dust, smoke and chemicals. The analysis was carried out on the 781 matched sets for which adequate information was available. Occupational exposure to paints in a full-time job for at least six months, eight to 28 years before diagnosis was not associated with an increased risk for bladder cancer in men (age-adjusted RR, 1.1; 95% CI, 0.77-1.6; 204 cases) but it was for women (RR, 3.9; 0.9-26.7; 14 cases). Little difference in risk was seen between commercial (RR, 0.90; 95% CI, 0.39-2.1; 49 cases) and spray (RR, 0.91; 95% CI, 0.48-1.7; 67 cases) painting in men. The authors noted the problems associated with the very low response rate, the inclusion of cases with borderline malignancies and the potential for recall bias.

In the study of Siemiatycki *et al.* (1987a,b) (see p. 70), an increased risk for bladder cancer was seen among people exposed to white spirits, 21% of whom worked in construction trades, mostly comprising painting.

(v) *Cancer of the biliary tract*

Cases of biliary tract cancers were identified from the National Swedish Cancer Registry for the period 1961-79, and the occupations of the patients identified from the 1960 census of occupations (Malker *et al.*, 1986). SIRs were calculated using the incidence rates for the total population and data from the 1960 census with regard to occupation and industrial employment, adjusted by region as well as by age and sex. There were 1304 cases of gall-bladder cancer and 764 cases of other biliary tract cancers in men, and 947 and 346 cases, respectively, in women. Significant SIRs of 1.3 (32 cases) and 1.4 (19 cases) for male painters and paperhangers were reported for gall bladder and other biliary cancers, respectively.

(vi) *Cancer of the pancreas*

Norell *et al.* (1986) reported on both a case-control study of pancreatic cancer and a retrospective cohort study of workers based on registry data in Sweden during 1961-79. Information on occupation was obtained through questionnaires. The case-control study in-

cluded 99 cases of pancreatic cancer (aged 40–79) and 163 hospital controls of the same age and sex with inguinal hernia and 138 population controls of the same age, sex and residence. A significant excess risk was seen for exposure to paint thinners (ten cases; RR *versus* population controls, 2.5; 90% CI, 1.1–5.9; RR *versus* hospital controls, 1.4; 90% CI, 0.7–2.9). In the cohort study, a 20% excess of pancreatic cancer was seen in workers (aged 20–64) in paint and varnish factories (90% CI, 0.7–1.9) and a 30% excess for floor polishing (90% CI, 0.6–2.3).

(vii) *Haematopoietic neoplasms*

Studies on leukaemia are summarized in Table 26.

In a case–control study of leukaemia in three geographical areas of the USA in 1959–62, information was collected on occupations and other subjects by personal interview (Viadana & Bross, 1972). The controls were a random sample from households in the area matched for age and sex. The analysis was limited to 1345 adult leukaemia cases and 1237 adult controls in whites. No association was seen between any occupation and leukaemia in women. The risk for leukaemia in men appeared to be associated with work in the construction industry, and specifically with painting. The risk for painters *versus* nonpainters was 2.8 [1.4–6.0]; that in comparison with clerks was 3.1.

Timonen and Ilvonen (1978) interviewed 45 adults in northern Finland with acute leukaemia or chronic myeloid leukaemia between 1973–77 and a control group of 45 patients from the same hospital about use of drugs and chemicals, including paint. Four cases and four controls had been exposed to paint containing benzene derivatives and lead.

Flodin *et al.* (1986) performed a case–control study on acute myeloid leukaemia on 59 cases aged 20–70 years in 1977–82 from five hospitals in Sweden. Living patients and controls replied to a questionnaire about solvent exposure. Two series of controls were used: 236 matched for sex, age and residence, and 118 selected randomly from the same general population. For ‘solvents, all kinds’, there were 11 cases exposed and 58 controls (crude rate ratio, 1.2); no case but five controls were classed as ‘painters’.

A population–based case–control study of 125 adult leukaemia cases and an equal number of controls matched for age, sex and residence was performed in Sweden in 1980–83 (Lindquist *et al.*, 1987). Information on occupation was obtained by a standardized questionnaire. ‘Painters’ included spray painters, car painters, machine painters, boat painters, asphalt painters and building painters. Thirteen cases and one control had been painters (RR, 13.0; 95% CI, 2.0–554). The median duration of exposure for painters was 16 years. After exclusion of case–control pairs with a ‘painter’, 26 patients and seven controls had worked in occupations which also involved exposure to paint and/or solvents and/or glues (RR, 3.7; 95% CI, 1.6–10.1).

Linnet *et al.* (1988) linked records for Swedish men by major industry and occupational categories from the 1960 census to cancer registry data for 1961–79 to calculate SIRs for leukaemia subtypes. Expected numbers were based on a 19-year follow-up, taking account of age, region and birth cohort. Among men classified as painters or paperhangers, SIRs were 1.1, 1.0, 1.1 and 0.8 for acute lymphocytic, chronic lymphocytic, acute nonlymphocytic and chronic myelocytic leukaemia, respectively (based on three, 41, 33 and 14 cases, respectively).

Table 26. Case-control and other studies of leukaemia among persons exposed in paint manufacture and painting

Reference	Location, time	Type of controls	Source	Exposure	No. of cases (no. of painters) ^a	RR	95% CI	Comments
Viadana & Bross (1972)	USA, 1959-62	Population	Interview	Painter	845 men (31)	2.8	[1.4-6.0]	In comparison with non-painters
						3.1	NA	In comparison with clerks
Timonen & Ilvonen (1978)	Finland, 1973-77	Hospital	Interview	Paint containing benzene derivatives and lead	45 adults (4)	1.0	-	
Flodin <i>et al.</i> (1986)	Sweden, 1977-82	Population	Interview	Painter	59 adults			
Lindquist <i>et al.</i> (1987)	Sweden, 1980-83	Population	Interview	Painter	125 adults (13)	13	2.0-554 1.6-10.1	Adjusted for other exposures
				Other professions exposed to paint and/or solvents and/or glues		3.7		
				Daily exposure to organic solvents (white spirits) and gasoline		3.0		
				Organic solvent		2.0		
	Petroleum products	1.4	(significant)					
Linet <i>et al.</i> (1988)	Sweden, 1961-79	Record linkage registry to census (cohort)	1960 Census record	(91)				
				Acute lymphocytic (3)		SIR, 1.1		
				Chronic lymphocytic (41)		SIR, 1.0		
				Acute nonlymphocytic (33)		SIR, 1.1		
				Chronic myelocytic (14)		SIR, 0.8		

A total of 25 cases of Hodgkin's disease in men aged 20–65 was studied in 1978–79 using two controls selected from the Swedish population registry (Olsson & Brandt, 1980). Subjects were asked about occupations, and occupational exposure was defined as handling organic solvents every working day for at least one year within the closest ten-year period. There was a significant association between Hodgkin's disease and exposure to solvents (12 cases; RR, 6.6; 95% CI, 1.8–23.8). Three of the 12 cases and only one of six controls exposed to solvents were painters (RR, 1.7; [0.09–54.6]); the RR for painters among all subjects was 6.7 [0.56–177.0]. [See also the monograph on some petroleum solvents.]

Vianna and Polan (1979) studied mortality in 1950–69 from reticulum-cell sarcoma, lymphosarcoma and Hodgkin's disease among 14 occupational groups considered to be exposed to benzene and/or coal-tar fractions in New York State. The exposed populations were estimated from census data, and deaths were obtained from health department records; mortality, adjusted for age, was presented separately for each cancer site and compared with rates for the state. Among 21 951 painters, the SMR for reticulum-cell sarcoma was 110 (based on nine cases), that for lymphosarcoma, 97 (15 cases) and that for Hodgkin's disease, 135 (21 cases).

Friedman (1986) carried out a case-control study of multiple myeloma among members of the Kaiser Foundation Health Plan in California, USA, and identified 327 cases during the period 1969–82. These were matched by sex, age, race, date of enrollment and residence with 327 controls on the rolls at the time of case diagnosis. Information on occupation was obtained from medical records. Painters as an occupational group occurred more frequently among cases (6) than controls (2). [The Working Group noted that it was not stated how frequently information on occupation was available.]

Morris *et al.* (1986) conducted a multicentre population-based case-control study in the USA of 698 newly diagnosed cases of multiple myeloma aged under 80 during 1977–81 and 1683 neighbourhood controls matched by age, sex and race. In personal interviews with subjects themselves or with next-of-kin, exposures were ascertained through a question about any exposure to toxic substances. A toxicologist grouped exposures into 20 categories, including 'paints, paint-related products and/or other organic solvents', which resulted in a RR adjusted for age, sex, race and study centre of 1.6 (51 cases exposed to paints and/or solvents; 95% CI, 1.1–2.4); of these cases, 40 had been exposed to paints and paint-related products. This risk showed little variation according to time since first exposure. When only cases who had been interviewed themselves were included, the adjusted RR for paints and/or solvents was 1.8 (39 cases; 95% CI, 1.2–2.7). [The Working Group noted that there may have been bias in the reporting of exposure.]

A case-control study of multiple myeloma in six areas of England and Wales was carried out by Cuzick and De Stavola (1988). A total of 399 cases identified at major regional centres between 1978 and 1984 and 399 age- and sex-matched hospital controls were interviewed about their past occupation and exposure to chemicals and radiation, as well as prior and family history of disease and immunizations. The risk of multiple myeloma in painters, including spray painters, was 1.9 (15 exposed cases; [95% CI, 0.76–4.7]).

Olsson and Brandt (1988) reported a case-control study of 167 male cases of non-Hodgkin's lymphoma aged 20-81 seen in the oncology department of the University Hospital of Lund, Sweden, in 1978-81. Exposure was assessed by interview by one of the authors using a standardized questionnaire, as in the study of Olsson and Brandt (1980; see p. 417). Two control groups comprising a total of 130 men who had been interviewed for two other case-control studies were used to estimate the exposure frequency. The RR for 'organic solvents' was 3.3 (63 exposed cases; 95% CI, 1.9-5.8). The risk for supradiaphragmatic lymphoma was higher (RR, 3.4; 95% CI, 2.3-5.2) than that for lymphomas localized below the diaphragm (RR, 1.4; 95% CI, 1.0-2.0). The risk increased with duration of solvent exposure. Occupational exposure to solvents was associated with employment in machine shops, chemical industry, painting, printing, wood industry and many other types of work; 14% of this population were painters.

(viii) *Cancer of the prostate*

In a cancer registry-based case-control study in Missouri, USA, conducted by Brownson *et al.* (1988), 1239 cases of histologically confirmed prostatic cancer in white males diagnosed between July 1984 and June 1986 were compared to 3717 white male cancer controls diagnosed in the same time period and frequency-matched by age. Information on occupation, collected routinely using a standardized protocol in all hospitals, was coded at the Registry as usual occupation and industry using the 1980 US census codes. When compared to workers in 'low-risk' industries (wholesale and retail trade, finance, insurance, real estate, business services and professional services), an elevated age-adjusted RR for prostatic cancer was apparent for men whose usual industry was coded as manufacturing of paints and varnishes (five cases; RR, 5.7; 95% CI, 1.4-24.3). However, in the analysis of usual occupation, no risk for exposure to paint was seen. The authors recognized several study limitations, including the use of crude occupational information, multiple comparisons, and use of cancer patients as controls.

(ix) *Cancer of the testis*

Swerdlow and Skeet (1988) identified 2250 cases of testicular cancer from the South Thames Cancer Registry, UK, for the period 1958-77. The proportion of painters and decorators among cases was compared with that among controls with cancers other than those of the genital system or at an unspecified site and among controls with cancers sampled such that no site represented more than 15% of the cancers in an age group. Occupation was identified from the records for 75% of cases and 73% of controls. The risk for testicular cancer among painters and decorators was about half that in the comparison group of professional, technical workers and artists (RR, 0.45; 15 cases), for both seminoma (RR, 0.44) and teratoma (RR, 0.55).

(x) *Cancer of the nasal cavity*

Hernberg *et al.* (1983) conducted a case-control study of nasal and sinonasal cancers among cases collected from the cancer registers in Finland and Sweden and from hospitals in Denmark in 1977-80. The 167 cases in live patients who agreed to interview were matched by age, sex and country to controls with colon and rectal cancer. Many of the patients in the subgroup with lesions in the maxillary sinus were not interviewed. Exposures were coded by

an industrial hygienist on the basis of intensity, duration and time. Smoking histories were evaluated for the period ten years prior to diagnosis, and smokers were found to be more frequent among cases (54.5%) than among controls (45.5%); the investigators indicated only that snuff use was not an important risk factor. Exposure to paints and lacquers reportedly showed a strong association with nasal cancer, but the investigators indicated that exposure to wood dust was generally a confounding factor. Two cases and no control had been exposed only to lacquers and paints, and both cases had had other potentially carcinogenic exposures.

(d) *Cancer in children in relation to parental exposure*

Fabia and Thuy (1974) analysed data on paternal occupation for 386 children aged less than five years who had died from malignant disease in the province of Québec, Canada, in 1965–70, and for the 772 control children whose birth registrations immediately preceded and followed those births. Father's occupation at the time of birth, as reported on the birth certificate, was recorded; no specific occupation was given for 30 cases or 56 controls. For ten cases and 11 controls, the paternal occupation at birth was described as painter, dyer and cleaner, excluding other hydrocarbon-related occupations (RR, 2.0; 95% CI, 0.86–4.7) [the Working Group calculated that the RR among those whose father was a painter was 1.2 (eight cases; 95% CI, 0.42–3.6)]. Among the 218 children with leukaemia, five of the fathers were in this occupational group; among the 101 children with central nervous system tumours and the 25 with Wilms' tumour, one (1%) and none (0), respectively, of the fathers were in this occupational group.

Hakulinen *et al.* (1976) carried out a case-control study of all 1409 children under 15 years of age with cancer reported to the Finnish Cancer Registry during the period 1959–68. After excluding twins and cases for which the father's occupation was unobtainable, 852 cases were available for analysis. The child born immediately before the case in the same maternity welfare district was chosen as a control. Father's occupation recorded at the time of conception was compared for cases and controls. Father's occupation described as 'painter, dyer, printer' was recorded for 12 cases and 15 controls; leukaemia and lymphomas occurred in one case and six controls, brain tumours in five cases and three controls and other tumours in six cases and six controls.

Kwa and Fine (1980) carried out a case-control study of 692 children born in Massachusetts, USA, who died before the age of 15 during 1947–57 and 1963–67. Controls were chosen from among the children whose birth registration immediately preceded and followed that of the case subject, giving a total of 1384 controls. Father's occupation at the time of birth registration was described as 'painter, cleaner, dyer' for 10 cases and 24 controls, comprising seven leukaemias or lymphomas, one neurological cancer and one urinary-tract cancer.

Zack *et al.* (1980) interviewed the parents of 296 children with cancer attending the Texas Children's Hospital Research Hematology Clinic in Houston, TX, USA, from March 1976 to December 1977. Controls were chosen from among relatives of cases, from among children in the neighbourhoods where the cases lived and from among children who did not have cancer attending the same clinic (33% had haemostatic defects, 24% various anaemias and 23% nonhaematological disorders). Job history from the year before the birth of the child

until one year before cancer diagnosis was assessed by personal or telephone interview of a parent. The fathers of none of the cases were described as 'painter, dyer or cleaner'; the corresponding figures for fathers of controls were one for relatives, two for neighbours and one for children attending the same clinic. [The Working Group noted that the selection criteria were given for neither cases nor controls and that it was unclear whether information on exposures was obtained from mothers, from fathers or from both.]

Hemminki *et al.* (1981) described the paternal occupations of 2320 children aged 0–14 with cancer reported to the Finnish Cancer Registry in 1959–75, many of whom had been included in the study of Hakulinen *et al.* (1976). Controls were chosen from among children whose birth had been registered immediately before and immediately after that of the index child. Parental occupation was taken as that in the maternity welfare clinic records at the time of pregnancy. The overall RR for a father's occupation as painter was 1.4 ([95% CI, 0.67–2.9]; based on 40 discordant pairs); the odds ratio for leukaemia was 1.5 ([0.22–10.3]; based on 12 discordant pairs) and that for brain tumours was 2.6 ([0.70–9.6]; based on 14 discordant pairs). The excess of brain tumours was most marked for the more recent study period, 1969–75, in which a significantly elevated RR of 5.0, based on seven discordant pairs, was reported. Maternal occupation was recorded for 2659 children, but no data on mother's exposure to paint was presented. The authors noted that for the earlier period (1959–68) only 63% of the cases had been included in the analysis; but for 1969–75, 86% of cases were included.

In a case-control study (Peters *et al.*, 1981), cases of brain tumours in children under ten years of age at diagnosis in 1972–77 were identified from the Los Angeles County Cancer Surveillance Program. Controls were matched to each case by sex, race and year of birth; matching for social class was attempted by trying to locate the control from among friends of the case or from the same neighbourhood. Mothers of 98 cases (84% of those available) and of 92 controls were interviewed by telephone, and the 92 matched pairs were analysed. Information included working and exposure histories of the mother and father before the pregnancy, during the three trimesters of pregnancy, during nursing and at the time of diagnosis. The authors noted the possibility of biased reporting and recording of exposures. Seven fathers of cases were reported to have had exposure to paints at any time from one year before conception up to the time of diagnosis; the father of one control had been similarly exposed. [The Working Group noted that this study addressed any exposure to paints and not only occupational exposures.]

Sanders *et al.* (1981) studied 6920 children under the age of 15 years who had died of malignant disease in England and Wales in 1959–63 and 1970–72. Father's occupation reported on the child's death certificate was compared with that recorded on the death certificate for a total of 167 646 childhood deaths that had occurred during the same periods. The PMRs for father's occupation described as 'painter or decorator' were 97 (based on 93 cases of cancer) in 1959–63 and 74 (based on 34 cases) in 1970–72. [The Working Group noted that data on specific cancer sites were not given.]

Associations between paternal occupation and childhood leukaemia and brain tumours were investigated in a case-control study in Maryland, USA (Gold *et al.*, 1982). Patients un-

der the age of 20 with leukaemia (1969–74) or brain tumours (1965–74) were ascertained in the Baltimore Standard Metropolitan Statistical Area from death certificates and records from 21 of 23 Baltimore hospitals. Two control groups consisted of children with no malignant disease, selected from birth certificates at the Maryland State Health Department, and of children with malignancies other than leukaemia or brain cancer. Information on occupational exposures of both parents before the birth of the child and between birth and diagnosis was collected by interviewing the mother. A total of 43 children had leukaemia and 70 had brain tumours. Paternal occupational category 'painter' was reported for one case of leukaemia, compared with three normal controls and none of the cancer controls, and no case of brain tumour, compared with one case in normal controls and none in cancer controls.

Wilkins and Sinks (1984) carried out a case-control study of 62 children with Wilms' tumour identified between 1950 and 1981 at the Columbus (Ohio) Children's Hospital Tumor Registry for whom paternal occupation was available from the child's birth certificate. Two groups of controls were chosen from birth certificates, the first matched individually for sex, race and year of birth, and the second for sex, race, year of birth and mother's county of residence at the time of the child's birth. Three of the fathers of cases were reported to be painters compared to one and none in the two sets of controls.

Van Steensel-Moll *et al.* (1985) carried out a case-control study of 713 children under 15 years of age with leukaemia diagnosed between January 1973 and January 1980 in the Netherlands. Controls were chosen from census records, matched by region, sex and age (to within two months). Information on occupational and other exposures of both parents during pregnancy was obtained by postal survey; the response rate was 88% for parents of cases and 66% for those of controls. The analysis was restricted to 519 patients with acute lymphocytic leukaemia and 507 controls. Twenty-five mothers of children with leukaemia and 11 mothers of controls reported having had occupational exposure to 'paint, petroleum products or other chemicals' during pregnancy (RR, 2.4; 95% CI, 1.2–4.6). These exposures were reported by 140 fathers of children with leukaemia and 113 fathers of controls (1.2; 0.8–1.7). The RR for paternal occupation described as 'painter, cleaner or dyer' was 1.6 (0.5–5.0) for exposures during pregnancy (eight cases) and 1.3 (0.4–4.0) for such exposures one year before the diagnosis of leukaemia (eight cases).

Lowengart *et al.* (1987) reported a case-control study of 123 children aged ten years or under with leukaemia identified in the Los Angeles County Surveillance Program in 1980–84, representing 57% of eligible cases. Controls were selected from among friends of cases or by random-digit dialling. Interviews were carried out by telephone and included questions on exposure to paints or pigments before, during and after pregnancy and on experiences the children had had from birth to the reference date. The specific types of exposure included in the general category 'paints or pigments' were spray paints, other paints, dyes or pigments, printing inks and lacquers or stains. Excess risks were observed for exposure of fathers to spray paints during the pregnancy (RR, 2.2; [95% CI, 0.91–5.3]) and after the pregnancy (2.0; 0.96–4.4) and for exposure to dyes and pigments during the pregnancy (3.0; [0.41–2.2]) and after the pregnancy (4.5; 0.93–42.8). The RR associated with 'spray paints' or 'dyes or pigments' was higher (RR, 2.5) if the father's exposure had been frequent (≥ 50

times per year) than if it had been less frequent (< 50 times per year; RR, 1.8) after the birth of the child. Data on maternal occupational exposure were not presented. Use of paints or lacquers in the home by the mother and/or father during the pregnancy and lactation gave a RR of 1.4 (0.79–2.6). [The Working Group noted that the exposure categories overlapped.]

Johnson *et al.* (1987) analysed paternal occupational exposures recorded on the birth certificate of 499 children aged 0–14 who had died of an intracranial or spinal cord tumour in Texas in 1950–79. Children who had been born outside Texas were excluded. Controls were chosen from a 1% sample of live births in Texas during the same period. Maternal occupation could not be assessed. A RR of 1.0 (95% CI, 0.3–3.3) was reported for paternal occupation described as a painter.

4. Summary of Data Reported and Evaluation

4.1 Exposures

Approximately 200 000 workers worldwide are employed in paint manufacture. The total number of painters is probably several millions, a major group being construction painters. Other industries in which large numbers of painters are employed include manufacture of transportation equipment and metal products, automotive and other refinishing operations and furniture manufacture.

Thousands of chemical compounds are used in paint products as pigments, extenders, binders, solvents and additives. Painters are commonly exposed by inhalation to solvents and other volatile paint components; inhalation of less volatile and nonvolatile components is common during spray painting. Dermal contact is the other major source of exposure. Painters may be exposed to other chemical agents that they or their coworkers use.

Painters are commonly exposed to solvents, the main ones being petroleum solvents, toluene, xylene, ketones, alcohols, esters and glycol ethers. Chlorinated hydrocarbons are used in paint strippers and less frequently in paint formulations. Benzene was used as a paint solvent in the past but is currently found in only small amounts in some petroleum solvent-based paints. Titanium dioxide and chromium and iron compounds are used widely as paint pigments, while lead was used commonly in the past. Asbestos has been used as a paint filler and may occur in spackling and taping compounds; painters in the construction industry and shipyards may also be exposed to asbestos. Exposure to silica may occur during the preparation of surfaces in construction and metal painting.

Workers in paint manufacture are potentially exposed to the chemicals that are found in paint products, although the patterns and levels of exposure to individual agents may differ from those of painters. Construction painters may be exposed to dusts and pyrolysis products during the preparation of surfaces and to solvents in paints, although water-based paints have become widely used recently. In metal and automobile painting, metal-based antirust paints and solvent-based paints are often applied by spraying; in addition, newer resin systems, such as epoxy and polyurethane, are commonly used. In contrast to other

painting trades, furniture finishing involves the use of more varnishes, which have evolved from cellulose-based to synthetic resin varnishes, including amino resins which may release formaldehyde.

4.2 Human carcinogenicity data

The reports most relevant for assessing the risk for cancer associated with occupational exposures in paint manufacture and painting are three large cohort studies of painters and collections of national statistics on cancer incidence and mortality in which data on cancer at many sites were presented for painters. These show a consistent excess of all cancers, at about 20% above the national average, and a consistent excess of lung cancers, at about 40% above the national average. The available evidence on the prevalence of smoking in painters, although limited, indicates that an excess risk for lung cancer of this magnitude cannot be explained by smoking alone. The risks for cancers of the oesophagus, stomach and bladder were raised in many of the studies, but the excesses were generally smaller and more variable than those for lung cancer. Some of the studies also reported excess risks for leukaemia and for cancers of the buccal cavity and larynx.

Several other small cohort and census-based studies in painters provided estimates of risk for cancer at one or several sites. The risk for lung cancer was reported to be raised in eight, that for stomach cancer in two, that for bladder cancer in two, that for leukaemia in four, that for malignancies of the lymphatic system in three, that for buccal cancer in three, that for laryngeal cancer in one, that for skin cancer in one, and that for prostatic cancer in three. In many studies, risks for cancer were reported only for sites for which the result was statistically significant.

In the three cohort studies of workers involved in the manufacture of paint, two of which were small, there was little to suggest an excess risk of lung cancer or of cancer at any other anatomical site.

Eleven case-control and related studies of lung cancer could be evaluated. All of the studies showed an increased risk for lung cancer among painters. The five studies in which smoking was taken into account showed an increase of 30% or more in risk for lung cancer. Two studies suggested increased risks among painters for laryngeal cancer, and one indicated an increased risk for mesothelioma.

Cancer of the urinary tract has been examined in relation to exposure to paint in 15 case-control and related studies. Eight showed an excess risk for bladder cancer in all painters. In certain studies, specific aspects of exposure to paint were examined: car painters were addressed in two studies, one indicating an excess risk; spray painters were evaluated in three studies, two of which showed an excess risk; and exposure to lacquer and chromium was associated with a risk in one study.

In a study of occupational histories of patients with oesophageal and stomach cancers, high risks were seen for painters. A further study also identified a risk for stomach cancer and another a risk for oesophageal cancer. One study of cancer of the gall-bladder and of the biliary tract showed associations with the occupation of painting. A study of pancreatic cancer reported a high risk for exposure to paint thinners.

Five studies of leukaemia mentioned painters. Two studies showed excess risks. Two small studies of Hodgkin's disease and three studies of multiple myeloma showed increased risks in association with the occupation of painter or with any exposure to paints, paint-related products or organic solvents.

A single study of prostatic cancer showed a significant excess risk for manufacturers of paints and varnishes, and one study reported a high risk for testicular cancer among spray painters.

Twelve studies of childhood cancer mentioned paternal exposure to paint and related substances; four of these also presented data on maternal exposure. Three studies showed an excess of childhood leukaemia in association with paternal exposure and one in association with maternal exposure. Two studies showed an excess risk for brain tumours in the children of male painters. One small study of children with Wilms' tumour showed an excess in those whose fathers were painters. All of these excesses are based on small numbers of children whose parents had been exposed, even in the larger studies. In the other studies, no association was seen between parental exposure to paint and childhood cancers. The type and timing of exposure varied among these studies.

4.3 Other relevant data

Painters may suffer from allergic and nonallergic contact dermatitis, chronic bronchitis and asthma, and adverse effects on the nervous system. There is also some indication of adverse effects in the liver, kidney, blood and blood-forming organs. Many of these effects are also seen in paint production workers.

Of three studies on the fertility of painters, two showed no adverse effect and the third a possible excess frequency of infertility in men. One study reported an excess frequency of spontaneous abortion in female painters, based on self-reported data. Studies of birth weight, perinatal mortality rates and congenital malformations in the offspring of male painters generally showed no adverse effects; few data on female painters were available.

No increase in the frequency of sister chromatid exchange in peripheral lymphocytes was found in one study of painters or in one study of paint manufacturing workers.

4.4 Evaluation¹

There is *sufficient evidence* for the carcinogenicity of occupational exposure as a painter.

There is *inadequate evidence* for the carcinogenicity of occupational exposure in paint manufacture.

Overall evaluation

Occupational exposure as a painter *is carcinogenic (Group 1)*.

¹For definitions of the italicized terms, see Preamble, pp. 27-30.

Occupational exposure in paint manufacture is not classifiable as to its carcinogenicity (Group 3).

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