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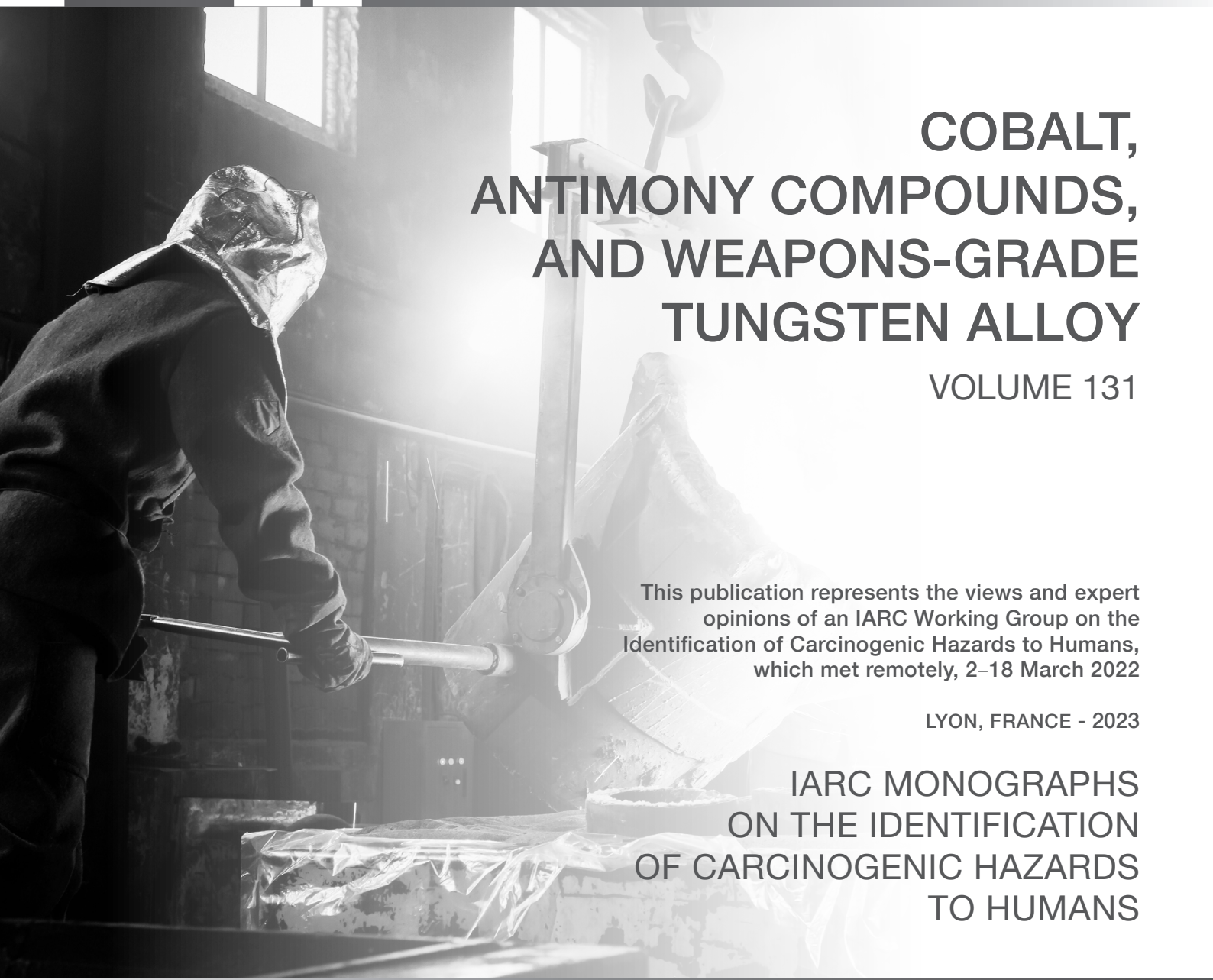

COBALT, ANTIMONY COMPOUNDS, AND WEAPONS-GRADE TUNGSTEN ALLOY

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International Agency for Research on Cancer

World Health Organization

Table S1.16 Exposure assessment review and critique for epidemiological studies on cancer and exposure to trivalent and pentavalent antimony

Reference and outcome	Agent	What was the study design?	What methods were used for the exposure assessment (including data source, environmental and biological measurements etc.)?	What was the exposure context?	Was exposure assessment qualitative, semiquantitative, or quantitative?	Concerns noted on sampling and collection protocols for metal measurement	What routes of exposure were assessed?	What exposure metrics were derived for use in analyses (e.g. average exposure, exposure duration, cumulative exposure etc.)?	What was the timing of exposure relative to the outcome?	Was there potential for co-exposures to other metals/carcinogens? If yes, were these accounted for in analyses?	Was there potential for differential or non-differential exposure misclassification?
Wingren & Axelson (1987) Stomach, colon, and lung cancer mortality	Antimony(III) oxide	Case–control	Employment information on death certificates supplemented by questionnaire from 7/13 glassworks in study area on past and present consumption of metals	Three alternative definitions investigated: (i) any glasswork employment; (ii) 6 categories of glassworkers by task (e.g. glass-blowers, grinders and etchers, polishers, packers etc.); and (iii) exposure categories according to metal consumption, based on glassworks questionnaires	Qualitative	No collection/measurement undertaken	Not quantified, but oral was implied for glassblowers	Only those categorical metrics described	"Past and present" employment recorded on death certificates; glassworks questionnaires collected information on present metals consumption and consumption occurring 25 yr before	Yes As noted by the authors, "analyses indicated very strong correlations between the use of many of the metals, and, consequently, it was impossible to separate the effects of isolated metals."; namely, glassworks using Pb (IARC Group 2A), As (IARC Group 1), and Sb were grouped in the analyses	Difficult to quantify, but possible as for other studies of this design; reference population consisted of other occupations, for which exposure to metals was not quantified
Wingren & Axelson (1993) Stomach, colon, and lung cancer mortality	Antimony(III) oxide	Case–control	Employment information on death certificates supplemented by questionnaire from 7/13 glassworks in study area on past and present consumption of individual metals: Sb, As, Cd, Cr, Cu, Pb, Mn, Ni, Se, Zn; a more detailed analysis of data used by Wingren and Axelson (1987)	Glassworkers who died in a parish where 1/7 glassworks was situated, which were categorized by annual consumption of individual metals: (i) no consumption; (ii) small amounts; and (iii) large amounts	Semiquantitative	No collection/measurement undertaken	Not quantified, but those applicable to glassworkers: oral and inhalation	Categorical level of metal consumption: none, low, and high	"Past and present" employment recorded on death certificates; glassworks questionnaires collected information on present metals consumption and consumption occurring 25 yr before	Yes Potential for co-exposures to 9 other metals that were quantified in the same manner as Sb; high correlation between metals and particularly between Sb and Pb (0.76); adjustment seemingly not undertaken	Yes: as applicable to categorical exposure assessment of this design
Kotsopoulos et al. (2012) *BRCA1*-related breast cancer incidence	Not specified	Case–control, unclear whether this is considered a nested study	Total Sb measured in fasting plasma samples with 13 other trace elements or micronutrients	Total Sb concentration in plasma	Quantitative: measurements were categorized into tertiles based on distribution among controls for regression analysis; trend tests made on continuous variables	Some quality control performance results not reported, e.g. reference material recoveries; it appears that the same internal standard – germanium – was used for all individual ICP-MS analytes irrespective of their differences in 1st ionization energy, not clear how this may have affected results	All relevant routes as reflected by plasma biomonitoring; study undertaken in a dietary/nutritional context	A single time-point concentration determined in plasma	While undertaken within a prospective study framework, cases with blood draw taken after diagnosis were included (79% of all cases) to increase case numbers; there were differences in the date of blood draw between cases and controls, later in cases by a mean of 2 mo	Measurements of 9 other metals (As, Ca, Cd, Cr, Cu, Fe, Mg, Se, Zn) were made; analyses were univariate; not possible to assess co-exposures due to study design and no information on exposure source; smoking status and alcohol consumption were not accounted for in analyses	Non-differential misclassification: possible (short half-life of Sb in plasma, single time-point samples, and differences in collection time within and between cases and controls)
García-Pérez et al. (2020) Incidence of cancer of the colon and rectum	Not possible to specify	Case–control	Participant's current residence was geocoded and used to calculate shortest distances from 134 point sources of industrial pollution reporting releases, including Sb, to the European Pollutant	Residential proximity to industrial point sources	Qualitative proxies of exposure and quantitative	N/A, metals not directly measured	Routes relevant to industrial pollutant releases to water, air, and soil	Dichotomous classification of distance between residence within 1, 1.5, 2, 2.5, or > 3 km and: 1) any industrial facility, 2) any industrial facility within specific industrial groups (n = 22), 3) facilities	To account for an estimated 10 yr latency of tumours of the colon and rectum, facilities that came into operation before the 10 yr before the mid-year of recruitment were included	Yes Potential for co-exposures to other pollutants released from these industrial plants; for classification 4 (specific pollutants), analyses did not adjust for other possible pollutants; tobacco smoking and alcohol consumption	Differential misclassification: unlikely Non-differential misclassification: likely

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			Release and Transfer Register in 2009					releasing specific groups of compounds (e.g., metals), and 4) facilities releasing specific pollutants (Sb)	Emissions data were used for 2009, which falls midway in the period over which cases and controls were identified (2008–2013)	were accounted for in the analyses	
García-Pérez et al. (2021) Gastric cancer incidence	Not possible to specify	Case–control	The same methodology as García-Pérez et al. (2020) was used	See García-Pérez et al. (2020)	See García-Pérez et al. (2020)	See García-Pérez et al. (2020)	See García-Pérez et al. (2020)	See García-Pérez et al. (2020)	See García-Pérez et al. (2020)	See García-Pérez et al. (2020)	See García-Pérez et al. (2020)
Liu et al. (2021) Thyroid cancer incidence	Not possible to specify	Case–control	Total Sb measured in urine samples (after 8 h fast) with 11 other elements	Total Sb concentration in urine corrected for specific gravity	Quantitative: measurements were categorized into quartiles for regression analysis; trend tests made on continuous variables		All relevant routes as reflected by urinary biomonitoring, but not specified	A single time-point concentration determined in urine	Urine samples were collected after recruitment, i.e. after diagnosis in cases	Measurements of 9 other metals (Cr, Mn, Co, Ni, Mo, Cd, Hg, Tl, and Pb), As, and Se in urine were made; some models were multiple-element; single-element model was adjusted for smoking and alcohol drinking; lack of information on sources of exposure limits interpretation of co-exposures	Non-differential misclassification: possible (short half-life of Sb in urine, single time-point samples, collection time (post-diagnosis), and lack of information on source of exposure)
Kresovich et al. (2019) Breast cancer incidence Tumour estrogen and progesterone receptor status	Not intended to be specified (population study)	Case series [case–case comparison]	Geocoded residential addresses of participants in 2002 were linked to census tract-level data on annual levels of metals in outdoor air from the 2002 US EPA NATA data	Annual census tract estimates of Sb concentrations in outdoor air linked to baseline residences among women participating in a population-based study of 696 women with a breast cancer diagnosis (2005–2008) from the Breast Cancer Care in Chicago (BCCC) study	Quantitative	None	Inhalation	Sb in air (ng/m^3)	Prior to the outcome	The NATA data contain modelled estimates for other metals (As, Be, Cd, Co, Cr, Pb, Mn, Hg, and Ni) and Se that were considered in the analyses (principal component analysis)	Differential misclassification: unlikely Non-differential misclassification: likely because census tract-level concentrations are proxies for personal exposures, methods did not account for changes in outdoor air levels over time, and there was no accounting for residential mobility
Mérida-Ortega et al. (2022) Breast cancer incidence	Not intended to be specified (population study)	Case–control, population-based	Total Sb measured in first void spot urine samples	Urinary Sb levels among 452 cases and 439 controls in northern Mexico recruited from 2007 to 2011	Quantitative	None	All routes	Sb in urine (µg/g creatinine)	Exposure was assessed after the outcome (for cases), but before treatment commenced	Urinary levels of other metals (Al, Cd, Cr, Ni, Pb, Co, Mo, Sn, and V) were measured and accounted for in the statistical analysis (principal component analysis); tobacco consumption and alcohol intake were accounted for in analyses	Differential misclassification: unlikely. Non-differential misclassification: likely due to the use of spot urine samples collected at baseline

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Binks et al. (2005) All-cause cancer mortality and specific-cause cancer mortality	Sn ore concentrates and residues were processed to produce (high-purity) Sn, Pb, Cu, Cd, Sb, and Ag	Retrospective cohort	Company's employment and pension records between 1 November 1967 and 28 July 1995 Death records from 1982 to 2001 (to determine person-time at risk)	Employment at an Sn smelter in North Humberside, United Kingdom for ≤ 12 mo between 1 November 1967 and 27 July 1995	Qualitative and quantitative	N/A	All routes (indirectly)	Employment (yes/no) For lung cancer mortality: Date of first employment Years of employment Years since entering employment Years since leaving employment	Prior to the outcome	Potential for co-exposures to other metals: Sn, Pb, As, Cd, radionuclides, and combustion products Exposure metrics were not specific to a particular contaminant	Differential misclassification: unlikely Non-differential misclassification: likely as time-related variables account for duration of employment but not magnitude
Duan et al. (2020) All-cause cancer prevalence and mortality	Not intended to be specified (general-population study)	Retrospective cohort	Concentrations of Sb in spot urine samples collected on participants from 1999 to 2014	Urinary levels of Sb among adults aged ≥ 20 yr participating in the US Centers for Disease Control and Prevention NHANES 1999–2014	Quantitative	None	All routes	Urinary (μg/L) concentrations (creatinine concentration was added as a covariate to the regression models) of Sb in a single spot sample at single point in time	Prior to the outcome	Potential for exposures to other metals and carcinogens; smoking by serum cotinine concentration Urinary levels of Ba, Cd, Cs, Co, Mo, Pb, Tl, W, and U were also measured in urine and evaluated, along with blood concentrations of Hg, Pb, and Cd The metal mixture was evaluated in WQS analyses, adjusted for smoking status (cotinine category)	Differential misclassification:-unlikely Non-differential misclassification: likely (use of a single urinary biomarker is not representative of long-term exposure and subject to substantial intra-individual variability (CDC, 2017; Wang et al., 2019a)
Guo et al. (2016) Self-reported, prevalent cancers All-cause cancer mortality	Not intended to be specified (general-population study)	Cross-sectional (prevalent outcomes) Retrospective cohort (mortality)	Concentrations of Sb in spot urine samples among NHANES participants, 1999–2010	Urinary levels of Sb among adults aged ≥ 18 yr participating in NHANES 1999–2010	Quantitative	None	All routes	Urinary concentrations of Sb (μg/g creatinine) in a single spot sample at single point in time	Exposures and outcome assessed at the same time in the prevalence study	Potential for exposures to other metals and other carcinogens No assessment of co-exposures in the analyses except for smoking and alcohol consumption	Differential misclassification:-unlikely Non-differential misclassification: likely (use of a single urinary biomarker is not representative of long-term exposure and subject to substantial intra-individual variability (Wang et al., 2019a))
Jones (1994) All-cause and specific-cause cancer mortality	Sulphide ore (containing 60% Sb and ≤ 0.5% As) to produce Sb metal, Sb alloys, and antimony(III) oxide	Prospective cohort	Unspecified (presumably plant personnel records)	Workers employed for ≥ 3 mo at an Sb plant in north-eastern England on 1 January 1961 and thereafter through 31 December 1992	Qualitative and quantitative	N/A	All routes (indirectly)	Assignment to 1/4 occupational groups: Sb workers; maintenance workers; zircon workers; others (including office workers and management staff), stratified by initial employment before and after 1 January 1961 For lung cancer mortality:	Prior to the outcome	Potential for exposures to other metals and carcinogens: As, As oxides, and PAHs Exposure metrics were not specific to a particular contaminant; the prevalence of smoking among workers in 1961 was 72%; smoking was not accounted for in analysis	Differential misclassification:-unlikely Non-differential misclassification: likely as time-related variables account for duration of exposure/employment but not magnitude

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Reference and outcome	Agent	What was the study design?	What methods were used for the exposure assessment (including data source, environmental and biological measurements etc.)?	What was the exposure context?	Was exposure assessment qualitative, semiquantitative, or quantitative?	Concerns noted on sampling and collection protocols for metal measurement	What routes of exposure were assessed?	What exposure metrics were derived for use in analyses (e.g. average exposure, exposure duration, cumulative exposure etc.)?	What was the timing of exposure relative to the outcome?	Was there potential for co-exposures to other metals/carcinogens? If yes, were these accounted for in analyses?	Was there potential for differential or non-differential exposure misclassification?
								Calendar period of first employment Years of exposure Years since first exposure			
Jones et al. (2007) Lung cancer mortality (same cohort as reported on in Binks et al., 2005)	Processed residues from other smelters and raw ores to produce pure Sb, pure Pb, Bi/Pb alloy, and Sb/Pb alloy	Retrospective cohort	Over 20 000 area and personal sampling measurements for Sb and other contaminants collected during 1972–1991 for estimation of cumulative exposures	Workers exposed to Sb in process and non-process areas at a United Kingdom Sb smelter over the period 1972–1991	Quantitative	Personal sampling data collected on a "campaign" basis; if this strategy was targeted towards "worst-case scenarios" then the exposure estimates are likely biased high Area samples collected as 15 min averages at set locations Unclear if there was any adjustment applied to the area sampling data that have been shown to underestimate personal exposures Breakdown in number of sampling results by contaminant was not provided No information on analytical protocols for analyses of area or personal samples	Inhalation	Cumulative exposure (mg-year/m^3) Cumulative exposures weighted for time since exposure and attained age	Prior to the outcome	Potential for exposures to other metals: As, Cd, Pb, and polonium-210 No assessment for co-exposures in the statistical analyses	Differential misclassification: unlikely Non-differential misclassification: likely, but difficult to quantify
Niehoff et al. (2021) Breast cancer incidence	Not intended to be specified (population-based study)	Case–cohort	Measurements of Sb in toenail cuttings collected at baseline from cohort participants enrolled from 2003 to 2009	Sb levels in toenail cuttings among women living in the USA and Puerto Rico aged 35–74 yr who enrolled in the Sister cohort study between 2003 and 2009	Quantitative	None	All routes	Concentrations (ng/g) of Sb in toenail cuttings collected at baseline, categorized into tertiles	Prior to the outcome	Potential for exposures to other metals and carcinogens Toenail concentrations of Al, As, Cd, Cr, Co, Cu, Fe, Mn, Mo, Ni, Se, Sn, and Zn were measured and evaluated A quantile-based g-computation approach was used to assess metal mixtures; analyses were adjusted for smoking status	Differential misclassification: unlikely Non-differential misclassification: likely (measurements in toenail cuttings of biomarkers collected at baseline is not representative of long-term exposure)
Schnorr et al. (1995) All-cause and specific causes of cancer mortality	Ore was composed of antimony oxides or antimony sulfide used to produce antimony(III) oxide	Retrospective cohort	Personnel and payroll records through 1975	Employment at an Sb smelter in South Texas, USA for ≥ 3 mo between 1 January 1937 and 1 July1971	Qualitative and quantitative	N/A	All routes (indirectly)	Employment (yes/no) For lung cancer mortality, stratification based on time since first exposure and duration of employment	Prior to the outcome	Potential for exposures to As Exposure metrics were not specific to a particular contaminant Mortality from smoking-related lung diseases was analysed but no smoking data on the cohort were available	Differential misclassification: unlikely

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White et al. (2019) Breast cancer incidence	Not intended to be specified (population-based study)	Prospective cohort	Modelled ambient air concentrations for Sb and Co using the 2005 US EPA NATA data linked to participants' geocoded residences at baseline, recruited from 2003 to 2009	Year 2005 census tract estimates of Sb concentrations in outdoor air linked to baseline residences among women participating in the Sister Study, recruited from 2003 to 2009	Quantitative	N/A	Inhalation	Census tract estimates of Sb exposure concentration (μg/m^3) linked to participants' residences at baseline, categorized into quintiles	Prior to the outcome	Potential for exposures to other metals and carcinogens including PAH and benzene levels and smoking Modelled estimates of As, Cd, Co, Cr, Pb, Mn, Hg, Ni, and Se were also evaluated WQS regression was used to assess metal mixtures; smoking status was accounted for in subgroup analyses	Differential misclassification: unlikely Non-differential misclassification: likely (due to the exposure assessment limited to residence at baseline, no accounting for variability in outdoor levels within census tracts, and variability in levels from year to year)

Ag, silver; Al, aluminium; As, arsenic; Ba, barium; Be, beryllium; Bi, bismuth; Ca, calcium; Cd, cadmium; Co, cobalt; Cr, chromium; Cs, caesium; Cu, copper; Fe, iron; Hg, mercury; ICP-MS, inductively coupled plasma mass spectrometry; JEM, job-exposure matrix; Mg, magnesium; min, minute; Mn, manganese; Mo, molybdenum; mo, month; N/A, not applicable; NATA, National Air Toxics Assessment; NHANES, National Health and Nutrition Examination Survey; Ni, nickel; OR, odds ratio; PAH, polycyclic aromatic hydrocarbon; Pb, lead; Sb, antimony; Se, selenium; Sn, tin; Tl, thallium; U, uranium; US EPA, United States Environmental Protection Agency; V, vanadium; W, tungsten; WQS, weighted quantile sum; yr, year; Zn, zinc.

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