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Assessment of human exposure to environmental sources of nickel in Europe: Inhalation exposure

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HIGHLIGHTS

- Local respiratory effects drive health risks of Ni via inhalation.
- DNELs for chronic inhalation 20 ng/m^3 (EU air guidance) and 60 ng/m^3 are considered.
- Most of the EU population is exposed to Ni air levels below the DNELs.
- A tiered modelling approach was used to assess Ni air levels near industrial sites.
- Majority of sites compliant with 60 ng Ni/m^3 (Tier I) or 20 ng Ni/m^3 (Tier I–IIb).

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ABSTRACT

The paper describes the inhalation nickel (Ni) exposure of humans via the environment for the regional scale in the EU, together with a tiered approach for assessing additional local exposure from industrial emissions. The approach was designed, in the context of REACH, for the purpose of assessing and controlling emissions and air quality in the neighbourhood of Ni producers and downstream users. Two Derived No Effect Level (DNEL) values for chronic inhalation exposure to total Ni in PM_{10} (20 and 60 ng Ni/m^3) were considered. The value of 20 ng Ni/m^3 is the current EU air quality guidance value. The value of 60 ng Ni/m^3 is derived here based on recently published Ni data (Oller et al., 2014). Both values are protective for respiratory toxicity and carcinogenicity but differ in the application of toxicokinetic adjustments and cancer threshold considerations. Estimates of air Ni concentrations at the European regional scale were derived from the database of the European Environment Agency. The 50th and 90th percentile regional exposures were below both DNEL values. To assess REACH compliance at the local scale, measured ambient air data are preferred but are often unavailable. A tiered approach for the use of modelled ambient air concentrations was developed, starting with the application of the default EUSES model and progressing to more sophisticated models. As an example, the tiered approach was applied to 33 EU Ni sulphate producers' and downstream users' sites. Applying the EUSES model demonstrates compliance with a DNEL of 60 ng Ni/m^3 for the majority of sites, while the value of the refined modelling is demonstrated when a DNEL of 20 ng Ni/m^3 is considered. The proposed approach, applicable to metals in general, can be used in the context of REACH, for refining the risk characterisation and guiding the selection of risk management measures.

1. Introduction

Nickel (Ni) is a natural element of the earth's crust. Natural sources like volcanic eruptions and windblown dust contribute to Ni in the atmosphere. Worldwide estimates of these natural emissions range from 8500 tons/year in early 1980s to 30,000 tons/year in the early 1990s (ATSDR, 2005). Additionally, anthropogenic activities like mining and smelting of Ni ores, manufacturing of Ni containing articles (e.g., stainless steel), fossil fuel combustion and waste incineration lead to supplemental emissions of Ni into the air. The anthropogenic emission rate to air is estimated to be a factor 1.4 to 1.8 higher than the natural one (IARC, 2012). Fossil fuel combustion is reported to be the major contributor of atmospheric Ni in Europe and the world, accounting for around 62% of the anthropogenic emissions in the 1980s (Barbante et al., 2002; ATSDR, 2005). Rydh and Svärd (2003) estimated that in

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1999 57,000 tons of Ni was released into the air from the combustion of fossil fuels worldwide. Other anthropogenic sources of Ni in the atmosphere are nickel smelting and refining processes which accounted for 17%, municipal incineration for 12%, steel production for 3%, other Ni-containing alloy production for 2% and coal combustion for 2% (ATSDR, 2005). For the EU27 total Ni emissions are estimated at about 1790 tons Ni/year of which 612 tons Ni/year is emitted to air. Traffic and industrial processes such as production (refining) and downstream use industries (metal surface treatment, production of batteries, etc.) have been identified as important sources for emissions of Ni to air (ECB, 2008; personal communication Patrick Van Sprang ARCHE).

Under REACH (Registration Evaluation Authorization & Restriction of Chemicals; EC 1907/2006), producers and importers of Ni and Ni substances, that are registered at >10 tons/year, have to demonstrate that manufacturing, selling, or using these substances in downstream applications has no adverse effects on human health and the environment. One aspect of the Chemical Safety Assessment (CSA) for REACH is the assessment and management of risks associated with indirect human exposure to each of the many environmental sources of Ni. The EU legislation requires an assessment of multimedia exposure for substances produced/used in the EU. Dietary Ni intake constitutes the main exposure pathway in terms of the total (absorbed) dose relevant for systemic health effects (De Brouwere et al., 2012). Inhalation of Ni is a minor source in terms of total daily doses relevant for systemic effects; however, there is also a concern about inhalation exposure to Ni due to possible adverse local effects on the respiratory tract.

This paper focuses on exposure and risk characterisation of Ni (as total Ni in ambient air) via the inhalation pathway. REACH requires a separate CSA for every substance that is registered in tonnage bands of 10 tons/year or above. However, although different chemical forms of nickel have different toxicological properties, a generic approach was used here because the speciation of emitted Ni substances and effects of atmospheric ageing on speciation of Ni particles is poorly understood. Thus, it is not possible to precisely apportion environmental Ni to its different chemical forms (Hughes et al., 1995). In general, however, oxidic and soluble forms of nickel predominate in ambient air (Füchtjohann et al., 2001; Galbreath et al., 2003; Tirez et al., 2011). In this manuscript a generic human inhalation exposure and risk characterisation approach was performed which is considered to be applicable to typical mixtures of Ni prevailing in ambient air, both at regional and local scale (i.e., near industrial sites).

The most critical adverse effects of Ni substances in rodent and/or human studies after inhalation are on the respiratory tract (e.g., lung inflammation, lung and nasal tumours) (e.g., ATSDR, 2005). A review of these data and a rationale for selecting the most appropriate points of departure to derive reference concentrations for Ni in ambient air based on respiratory tract effects are included in Oller et al. (2014).

The aims of the present study are i) to select appropriate Derived No Effect Level (DNEL) values to protect the general population from adverse respiratory effects associated with Ni exposure, starting from toxicity data and epidemiological data; ii) to characterise the regional EU ambient air exposure to Ni and to perform the risk characterisation for respiratory effects of Ni at the EU regional level; iii) to develop a tiered approach to aid industries in refining their local emission assessment; and iv) to perform risk characterisation at the local exposure level.

To fulfil these aims we undertook the following analyses: 1) a review of the most recently derived ambient air standards and their underlying assumptions, including the EU air guidance value which can be considered as both a DNEL (Derived No Effect Level) and a DMEL (Derived Minimal Effect Level) (see Results and discussions); 2) a derivation of an ambient air PM_{10} DNEL value that incorporates the concept of a practical threshold for the respiratory carcinogenicity (as well as toxicity) of nickel and includes full dosimetric adjustments; 3) a determination of the range of inhalation Ni exposure within the EU population (regional exposure assessment); 4) comparison of regional ambient air exposures to DNEL values (regional risk characterisation); 5) development of a tiered approach for assessing Ni inhalation exposure levels at local scales around Ni producing/using industries when measured data are lacking; and 6) demonstration of the applicability of this tiered approach to the risk characterisation of exposures in the Ni producing/using sectors (as an example of industrial Ni sites).

The approaches described in this study can be used in the context of fulfilling REACH requirements, both for nickel and for other metal substances to refine the risk characterisation when the screening approaches fail to demonstrate safe use. The applied models are not Ni specific and are valid for other substances defined as non-reactive in the atmosphere.

2. Materials and methods

2.1. Review of ambient air reference or guidance values for nickel

In considering the selection of a suitable DNEL to protect the general population from the lifetime effects associated with inhalation of Ni present in ambient air, a brief review of ambient air guidance or reference values derived in the last fifteen years within and outside the EU was undertaken.

2.2. Derivation of an ambient air PM_{10} DNEL value for nickel

In the last ten years, there has been growing recognition that nickel and other compounds may cause tumours by threshold driven pathways (Bolt and Huici-Montagud, 2008; Hengstler et al., 2003; SCOEL, 2011). In addition, more information on the toxicological effects of nickel compounds in rats and humans has become available (e.g., Oller et al., 2008; Goodman et al., 2009, 2011; Kraut et al., 2010) and the use of dosimetric models to refine the calculations of human equivalent concentrations (HECs) to the animal or workplace exposures have been more widely used (e.g., Maruyama et al., 2006). Recently, Oller et al. (2014) considered these issues in the derivation of Ni HECs corresponding to the PM_{10} aerosol subfraction of ambient air that can be used as modified points of departure for the derivation of a DNEL for nickel.

Since information on environmental exposure to different Ni species is not routinely collected, the nickel DNEL needs to cover all Ni forms present in ambient air. Ni monoxide and complex Ni–Fe oxides together with Ni sulphate predominate in ambient air, with minor contributions from Ni sulphides, and metallic nickel (e.g., Galbreath et al., 2003; Füchtjohann et al., 2001; Tirez et al., 2011; Oller et al., 2014). Both cancer and non-cancer respiratory effects are the main drivers for setting ambient air reference values for nickel. For respiratory toxicity effects, Oller et al. (2014) derived PM_{10} HECs based on rodent effects of Ni sulphate and Ni oxide but these HECs are protective for exposure to other Ni compounds as well. The authors applied toxicokinetic adjustments by considering equivalent retained doses in the pulmonary region of the respiratory tract in rats and in humans.

For cancer effects, Oller et al. (2014) and others (e.g., SCOEL, 2011; Bal et al., 2011; Goodman et al., 2011) considered that nickel compounds are likely to be indirect genotoxic carcinogens with a practical threshold. It is possible then to derive a nickel reference value for cancer that is based on a practical threshold identified in epidemiological cancer studies (i.e., a DNEL) instead of relying on a linear dose–response extrapolation and the acceptance of $1/10^5$ or $1/10^6$ extra cancer risk (i.e., a DMEL). Furthermore, exposure to soluble, oxidic and sulphidic Ni with focus on low sulphidic Ni exposure (thus mirroring Ni speciation in ambient air) can be considered in the epidemiological studies and dosimetric adjustments can be made to account for differences in particle size distribution between Ni refinery exposures and exposures in ambient air. This approach was taken by Oller et al. (2014) to derive a PM_{10} Equivalent Concentration (EC) corresponding to the practical threshold for lung cancer effects derived from epidemiological data presented in Goodman et al. (2011).