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## Balance sheet for extraction and use of limestone, and calculation of the pertinent CO<sub>2</sub> emissions

### Summary

by

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## 1 Introduction

As a Party to the United Nations Framework on Climate Change, Germany is obliged to prepare and regularly publish emissions inventories. For fulfilment of these reporting obligations, Germany maintains the "Central System of Emissions" (CSE), a comprehensive inventory database for calculation, administration and documentation of relevant emissions data. In connection with ongoing research efforts, the inventory database is continually reviewed, and gaps in its data are systematically identified and eliminated, with a view to inventory improvement.

The present research project (Implementation of the inventory plan, and national, independent review of emissions inventories for greenhouse gases – Sub-project 02 "Balance sheet for extraction and use of limestone, and calculation of the pertinent CO<sub>2</sub> emissions" ("Umsetzung des Inventarplanes und nationale unabhängige Überprüfung der Emissionsinventare für Treibhausgase - Teilvorhaben 02 'Bilanzierung der Gewinnung und Verwendung von Kalkstein in Deutschland und Ausweisung der CO<sub>2</sub>-Emissionen'") FKZ: 205 41 217/02) has contributed significantly to improvement of the national emissions inventory. In particular, in the area of emissions from raw materials, it has identified gaps in the data for thermal use of mineral carbonates (especially limestone). The aim of the research project was to eliminate gaps in the data for limestone use, and for the pertinent CO<sub>2</sub> emissions, within the CSE. The research project is important not only because it has improved and helped to assure the quality of the inventory for Germany; it has also closed gaps in emissions data for base year 1990, a significant achievement in that those data will be definitively defined in 2006, in connection with inclusion of data for the period through 2004. As of the end of the project, the scope of the relevant effort remains confined to the data through 2004. For this reason, the documentation required for updating the data as of 2005 will be provided to the Federal Environment Agency (UBA) as planned.

In this context, the Federal Environment Agency commissioned the Wuppertal Institute for Climate, Environment and Energy (WI), located in Wuppertal, and the EEFA research institute, located in Berlin and Münster, to identify all available empirical options for improving the inventory in the area of limestone use and to find practicable means of using such options in the CSE.

## 2 Concept and database

### 2.1 Conceptual and methodological aspects

In a first step, the research project identified the economic sectors in which limestone or lime is used and considered the significance of the emissions tied to the various uses. In this area, CO<sub>2</sub> emissions occur especially in production processes based on burning of limestone. At temperatures up to 1,150 °C, limestone, which consists mainly of calcium carbonate (CaCO<sub>3</sub>), reacts to form calcium oxide (CaO), via a deacidification process that releases CO<sub>2</sub>. CO<sub>2</sub> emissions from deacidification of carbonate-containing raw materials occur especially in high-temperature processes in the non-metallic minerals industry (cement industry, lime industry, bricks industry, etc.), as well as in the glass and steel industries, which also consume unburnt carbonate-containing raw

materials. The specific raw-material-related CO<sub>2</sub> emissions in the aforementioned production areas depend on the raw-material mixtures used – i.e., on the raw materials' CaCO<sub>3</sub> content.

In some production processes – such as processes in the sugar industry – producers operate their own shaft furnaces for burning lime. For its part, the sugar industry uses the lime (in the form of milk of lime), and the resulting carbon dioxide is bound, in subsequent processing, in carbonic acid. For this reason, limestone burning in sugar production may be considered CO<sub>2</sub>-neutral.

In addition, non-thermal limestone uses also cause CO<sub>2</sub> emissions. Important sources of CO<sub>2</sub> emissions from non-thermal limestone uses include use of limestone-containing fertilisers (liming of soil in the agricultural and forestry sectors) and flue-gas desulphurisation systems.

Finally, the research project sought to list, in detail, the CO<sub>2</sub> emissions occurring in emissions-relevant processes of limestone use that had not previously been included in the emissions inventory. In a first analysis step, limestone-production quantities and limestone uses were quantified, and physically based limestone balance sheets were prepared for the report years 1990 (base year), 1995 (supporting reference year that is used in connection with statistical transitions) and 2004 (current year).

To those ends, the research project used a concept that could best be described as a combination of a "bottom-up" approach and a "top-down" approach. The production side of the limestone balance sheet (i.e. limestone production, less exports, and plus imports) was prepared from the "top-down" side, primarily from official statistics and information available from relevant associations. In the area of limestone use, significant data restrictions apply. As a result, limestone inputs in a number of economic sectors (as defined by the deep-level sub-categorisation required for the present context) had to be determined via detailed individual calculations ("bottom-up").

To implement this "balance-sheet concept", the following was carried out:

- Relevant specific production conditions were analysed, with a view to identifying suitable indicators (for example, plaster production from flue-gas desulphurisation in power stations, production trends (in part, for individual production stages, and for preliminary products); the sulphur content of the fuels used; the chemical composition of the products produced (CaO fractions); or, directly, process emissions from limestone use) and
- Stoichiometric calculations, and data for specific limestone inputs (which were obtained from the literature), were used for the purpose of calculating total consumption of carbonate-containing raw materials.

Via this approach, the research project was able to compile and complete a large body of data that had previously been present only in widely scattered, incomplete form, and make it available for calculation of CO<sub>2</sub> emissions for the national emissions inventory.

The limestone balance sheet developed here can be used to highlight limestone substance streams, in a transparent manner, in order to reveal any double-counting of emissions within the inventory and any non-transparent structures in sub-areas of the inventory. Raw-material-related CO<sub>2</sub> emissions result only from use of unburnt carbonate-containing raw materials.

### **Terminology:**

In the interest of simplicity, in the following the term "limestone" is used even for cases in which limestone and dolomite are involved. Other carbonates are considered insignificant and thus not included, and raw materials that contain CaCO<sub>3</sub>, such as marl used for cement production, are not separately covered.

In this terminological framework, all relevant burnt products are referred to as "lime", for simplicity's sake. In some cases, an explicit distinction is made between burnt lime (from limestone) and dolomite lime (from dolomite stone).

In light of the great complexity of the area of limestone and lime use, the present research project has prepared both a limestone balance sheet and a lime balance sheet, along with an "auxiliary balance sheet".

In sum, the project focused on the following emphases:

- 1) Preparation of complete, consistent limestone and lime balance sheets for Germany. In an approach similar to that used for the energy balance, the overall area is divided horizontally, into the categories of production calculation, transformation balance sheet and final consumption (broken down by sectors). Vertically, the overall area is divided into the categories of limestone and lime. Complete balance sheets have been prepared for the three report years 1990, 1995 and 2004, in keeping with the requirements set forth in the relevant specifications.
- 2) Formation / calculation of complete times series for limestone use, broken down by economic branches / sectors for the period from 1990 until the current cut-off (2004). These time series can be linked, as separate limestone time series, with relevant key words in the CSE or linked with time series already present within the CSE. Apart from any considerations regarding implementation of the time series in the CSE, two relevant cases were differentiated: firstly, cases in which the sector's raw-material-related CO<sub>2</sub> emissions have already been taken into account, via production-oriented emission factors, in the CSE – and only the relevant limestone time series remains to be calculated or implemented; and, secondly, cases in which neither the sector's raw-material-related CO<sub>2</sub> emissions from limestone use nor its limestone consumption itself have been taken into account in the CSE (complete lack of data). In the interest of the consistency of emissions calculations, in the first case it must be remembered that the calculation approach used to derive the limestone time series must be compatible with the method already being used, in the CSE, to calculate raw-material-related CO<sub>2</sub> emissions.
- 3) Explicit description of CO<sub>2</sub>-relevant limestone quantities and, if necessary, calculation of the pertinent CO<sub>2</sub> emissions for the balance-sheet entries added to the inventory.

## 2.2 Empirical aspects and data sources used

As the research project's conceptual framework suggests, practical implementation of the project had to create significant demands with regard to availability of empirical data. The most important bases for calculation of limestone production are published in statistics of the Federal Statistical Office (Destatis) and of the Federal Institute for Geosciences and Natural Resources (BGR).

Domestic production of limestone and dolomite was determined primarily on the basis of official production statistics. Limestone production can be broken down into the following areas:

- Limestone for production of cement; burnt lime and limestone for industry and environmental technology applications (reporting no 1412 10 530)
- Limestone, ground (reporting no 1412 10 550)
- Dolomite, neither burnt nor sintered (reporting no 1412 20 530)
- Crushed limestone for structural concrete or as rock fill for road and railway construction (reporting no 1421 20 301)

There are many reasons why data for limestone extraction, as shown in production statistics, can differ from actual domestic production figures. First of all, the official production statistics for sector 14.21 include only operations with more than 10 employees, while the statistics for sector 14.12 include only operations with more than 20 employees. By its very nature, "production of non-metallic minerals" involves a high degree of mechanisation. Consequently, the productivity of employees in this area has tended to be higher than that of employees in other areas of the manufacturing sector. Therefore, there are likely to be numerous limestone-producing operations that achieve high production outputs with only small numbers of employees. On the other hand, no data on such operations, which are not covered by production statistics, are available, and thus it is not possible to derive precise figures for total limestone production in Germany from production statistics.

In many regards, the available data for limestone use is also incomplete. Source-based empirical figures for the German lime industry's sales of unburnt products are available for the period as of 1998. In the framework of the research project, the German lime industry association (Bundesverband der Kalkindustrie) provided sales statistics. Those statistics were not broken down in accordance with the categories required for the balance sheet, however (BV Kalk 2006b).

The data published by the German lime industry association cannot be precisely correlated with the economic sectors, from official statistics, whose limestone use is considered relevant with regard to CO<sub>2</sub> emissions.

Simply put, limestone demand at the level of individual economic sectors depends on production quantities, on the relative roles of different individual processes and on overall production structures (activity rates). The steel industry, for example, uses limestone only in processing of iron ores (sintering plants) and in pig iron production in blast furnaces. On the other hand, (burnt) steelworks lime is used – *inter alia*, as a slag former – in actual refining of raw steel in oxygen-steel or electric-steel processes. Wherever possible, the present research project has sought to describe sectoral

limestone-consumption quantities on the basis of technical (process-based) relationships. For calculation of limestone use in the steel industry, this means that limestone consumption is formally obtained as the product of specific limestone use (in kg per tonne) and pig-iron / sinter production (in tonnes). The advantage of this approach is that process-related limestone-use rates, at the level of specific production technologies (such as blast furnaces), are largely independent of economically based substitution processes (intrasectoral structural changes), and they can fluctuate only within narrow technically based ranges. Additionally, once the data sets obtained in this research project are added to the CSE, future required updating can be achieved via simple updating of data items (i.e. where data for specific limestone-consumption quantities is lacking), without any risk of major distortions.

The present research project has used a range of different data sources for calculation of limestone use on the demand side. The key production figures (activity rates) required, from the outset, for such calculation were derived on the basis of official production statistics (Produktion im Produzierenden Gewerbe ("Production in the manufacturing sector"), Fachserie ("specialised series") 4 Reihe (sub-series) 3.1) and on various surveys and statistics of relevant associations. In some cases, association statistics permit derivation of more complete production figures than do official statistics, since association statistics include operations/plants with fewer than 20 employees and since such publications – at least in part – support derivation of production data for all Germany, even for base year 1990.

On the other hand, figures for specific limestone consumption per product unit were obtained from technical publications (industry journals such as "Ziegelindustrie International" ("international bricks industry"), "Zement-Kalk-Gips" ("cement-lime-plaster"), etc.), from guidelines (for example, guidelines of the Association of German Engineers (VDI)), from experts of associations of industrial companies that use limestone and from stoichiometric calculations.

### **2.3 Relevant specific balance sheet entries (use side of the limestone balance sheet)**

Numerous balance-sheet entry categories were identified, in close co-operation with the entity that commissioned the study, and with responsible experts in the Federal Environment Agency, and then studied in the present research project. The various balance sheet entry categories differ in terms of their importance for calculation of the limestone balance sheet (and lime balance sheet). Similarly, the various balance sheet entry categories differ in terms of their importance for the existing emissions inventory. Table 2-1 shows all of the balance sheet entry categories in which limestone or lime is used and that were considered to be important in the context of the present research project.

The table also shows the balance sheet entry categories' emissions relevance. As the short outline of the relevant balance sheet entry categories shows, there is no "ideal" method for calculating limestone and lime use – i.e. a method that is valid for all source categories, conforms to the quality requirements for the emissions inventory, is empirically practicable and is universally applicable. In the present research project, derivations have thus always involved use of a mixture of different practicable calculation approaches. The calculation procedures used in relevant specific cases, and the pertinent results, are presented in detail, for each relevant economic sector, in the third chapter.

Table 2-1: Relevant balance sheet entry categories

| CRF    | Sector/name   | Input category    |     | Relevant in terms of CO <sub>2</sub> emissions? | Inventory completion in the framework of the project |
|--------|---|-------------------|-----|---|--|
|        |   | CaCO <sub>3</sub> | CaO |   |  |
| 1A1    | Electricity production, flue-gas desulphurisation   | x                 | x   | Yes   | Yes  |
| 2A1    | Cement production   | x                 |     | Yes   | Not necessary  |
| 2A2    | Lime production: burning of limestone and dolomite  | x                 |     | Yes   | Not necessary  |
| 2A4    | Soda ash production   | x                 |     | No  | Not necessary  |
| 2A7    | Glass production  | x                 |     | Yes   | Not necessary  |
| 2A7    | Bricks production (fraction in the raw meal)  | x                 |     | Yes   | Yes  |
| 2B4    | Calcium carbide production  |                   | x   | No  | Not necessary  |
| 2C1    | Iron and steel production   | x                 | x   | Yes   | Yes  |
| 2C3    | Clay production (for smelter aluminium)   |                   | x   | No  | Not necessary  |
| 2D1    | Paper industry  | x                 | x   | No  | Not necessary  |
| 2D2    | Sugar production  | x                 |     | No  | Not necessary  |
| 4D, 5D | Liming of soils in agriculture and forestry   | x                 | x   | Yes   | Not necessary  |
| 6B     | Water and sludge treatment  | x                 | x   | No  | Not necessary  |
|        | Other sectors (such as construction, other construction-materials industry and chemical industry, etc.) | x                 | x   | No  | Not necessary  |

Source: Own description

Annex 3 of this final report presents a schematic illustration of the limestone and lime balance sheet(s).

In the course of the study, balance sheet entry categories were found that have limestone or lime use but that lack reliable pertinent data that could be used to calculate the actual relevant limestone or lime consumption. This applies to the area of fertiliser production (2B5), for example. On the other hand, in that area limestone is simply used as an additive and is not emissions-relevant. The study did tally the applicable limestone quantity from fertiliser production for the entry category "Liming of soils in agriculture and forestry", where the limestone is emissions-relevant. Another area of limestone and lime use for which it was not possible to calculate input quantities, due to a lack of suitable data, is the balance sheet entry category Road paving with asphalt (2A6). The inputs in that balance sheet entry category are combined within the balance sheet entry category "Other sectors", and they are not emissions-relevant. For the balance sheet entry category Flue-gas desulphurisation, industry (1A2), lime inputs were calculated on the basis of the sulphur content of the fuels used and of the actual quantities of sulphur dioxide emitted. In addition, the chemical formula for the spray absorption process with hydrated lime was used. Due to a lack of precise data, that calculation procedure entails major uncertainties, however, and thus that entry category is not included within the balance sheet as such but instead is taken into account under "Other sectors".

## 2.4 Structure of the limestone balance sheet

When all limestone inputs, in all relevant economic sectors, are summed and then compared with relevant production, the limestone balance sheet shown in the following table results.

Table 2-2: Limestone balance sheet

| In millions of tonnes   | 1990   | 1995  | 2004  |
|---|--------|-------|-------|
| Domestic production   | 110,50 | 76,79 | 74,10 |
| Imports   | 0,13   | 2,28  | 2,71  |
| Exports   | 0,02   | 0,40  | 0,86  |
| Total available quantity (= domestic production + imports – exports)                                    | 110,61 | 78,66 | 75,96 |
| Use   |        |       |       |
| Lime industry   | 13,73  | 14,14 | 12,39 |
| Cement industry   | 34,20  | 35,13 | 31,83 |
| Soda ash production   | 2,27   | 1,83  | 1,70  |
| Glass   | 0,70   | 0,89  | 0,90  |
| Iron and steel  | 5,44   | 5,35  | 5,06  |
| Sugar   | 0,69   | 0,78  | 0,85  |
| Flue-gas desulphurisation, power stations   | 1,54   | 1,75  | 3,17  |
| Agriculture and forestry  | 2,44   | 3,23  | 3,15  |
| Water and sludge treatment  | 0,05   | 0,06  | 0,04  |
| Other sectors (such as construction, other construction-materials industry and chemical industry, etc.) | 49,54  | 15,49 | 16,88 |
| Total use   | 110,61 | 78,66 | 75,96 |

Source: Own description; cf. Annexes 4 and 5

In addition, the limestone fractions found in the raw meal for bricks production are taken into account in an "auxiliary balance sheet".

Table 2-3: "Auxiliary balance sheet", limestone fraction in raw materials

|                                       | Units              | 1990 | 1995 | 2004 |
|---------------------------------------|--------------------|------|------|------|
| Limestone in raw materials for bricks | Millions of tonnes | 1,0  | 1,4  | 0,9  |

Source: Own calculations

### 3 Summary

The present research project has been able to prepare a complete limestone / lime balance sheet for Germany. As a result, the commissioned research task has been completed. This involved identifying gaps in the data for thermal use of limestone – especially in the area of emissions from raw materials – and eliminating the gaps, in the area of limestone use and its pertinent CO<sub>2</sub> emissions, within the CSE.

The following table shows the balance sheet entry categories that are being added to the emissions inventory as of the 2006 report. These data include data determined, in the research project, for limestone use and for the limestone fractions in raw materials. They also include the Federal Environment Agency's conservative estimates for the area of flue-gas desulphurisation systems.

Table 3-1: Inventory additions for 2006: CO<sub>2</sub> emissions from limestone use

| CRF area | Name  | Units | 1990  | 1995  | 2004  |
|----------|---|-------|-------|-------|-------|
| 1A1      | Flue-gas desulphurisation systems in large combustion installations | kt    | 932   | 1.229 | 1.764 |
| 2A7      | Ceramics production, bricks production (fractions in raw materials) | kt    | 490   | 670   | 475   |
| 2C1      | Iron and steel<br>Pig iron<br>Sinter                                | kt    | 2.370 | 2.257 | 2.233 |

Source: NIR 2006 (including Errata 2C1)

As a result, the emissions for base year 1990, which are being definitively defined with the 2006 report, have been adjusted by some 3.5 million tonnes of carbon dioxide. This has enhanced the inventory's completeness.

Slight data recalculations in the further course of the project, following the 2006 report, have led to the inventory additions shown below. It should be noted that the relevant data will make their first (report) appearance in the 2007 report, where they (especially the data for the ceramics industry) will be subject to revision and updating by the Federal Environment Agency's responsible experts.

Table 3-2: Recalculations of CO<sub>2</sub> emissions from limestone use

| CRF area | Name  | Units | 1990  | 1995  | 2004  |
|----------|---|-------|-------|-------|-------|
| 1A1      | Flue-gas desulphurisation systems in large combustion installations | kt    | 679   | 769   | 1.394 |
| 2A7      | Ceramics production, bricks production (fractions in raw materials) | kt    | 452   | 609   | 409   |
| 2C1      | Iron and steel<br>Pig iron<br>Sinter                                | kt    | 2.392 | 2.354 | 2.225 |

Source: Own calculations for the conclusion of the project

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Annex „3“: balance sheets according to IPCC-source principle

