

Human health damage characterisation factors for particulate matter emissions to air for application in life cycle analysis

Technical Report

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Zusammenfassung

Die Belastung der Luft mit winzigen Staubteilchen ist eine der grössten Herausforderungen in der Luftreinhalte-Politik. Vor allem Städte und verkehrsnaher Gebiete leiden unter zu hohen Feinstaub-Belastungen. In diesen Gebieten liegen die Jahresmittelwerte für Feinstaub (=PM: Particulate Matter) über den geltenden Grenzwerten. Epidemiologische Studien weltweit konnten konsistent aufzeigen, dass diese Feinstaub-Belastungen beim Menschen zu einer Zunahme an Herz- und Atemwegserkrankungen führen. Feinstäube sind zusammengesetzt aus festen und flüssigen Anteilen und stammen aus verschiedenen Quellen: Verbrennungsprozesse, mechanischer Abrieb von Reifen und Strassenbelag, Abbauprozesse (z.B. von Gestein) etc. Die Verbrennung von Holzprodukten wird häufig als problematisch bezüglich Feinstaub-Belastung angesehen, ohne jemals eine objektive, ganzheitliche Bewertung zu Vor- und Nachteilen dieses Rohstoffes gemacht zu haben.

Die Methode der Ökobilanzierung (Life Cycle Assessment, LCA) wird oft eingesetzt um Umwelteffekte von Prozessen oder Produkten zu optimieren oder die Effekte verschiedener Prozesse oder Produkte miteinander zu vergleichen. LCA bietet einen Rahmen zur Identifizierung und Evaluation der Umweltauswirkungen (inkl. Auswirkungen auf den Menschen) über den gesamten Lebenszyklus (von der Rohstoffentnahme bis zur Entsorgung) von Produkten und Prozessen. So kann eine LCA potentielle Schäden an Gesundheit, Ökosystem und Ressourcen durch den Betrieb einer Holzfeuerung mit den potentiellen Schäden durch den Betrieb einer Öl-/Gasfeuerung bewerten, oder die Stückholzverbrennung kann mit der Verbrennung von Pellets verglichen werden. Voraussetzung dafür ist, dass Emissionen mit unterschiedlicher Wirkung in der LCA unterschieden werden können.

In der LCA-Praxis wird heute Feinstaub undifferenziert als PM₁₀ Masse (Feinstaub kleiner als 10 µm) bewertet. Es ist aber bekannt, dass es viele andere wichtige Partikeleigenschaften gibt, wie z.B. Grösse, Wasserlöslichkeit, chemische Toxizität, welche die Wirkungen auf den Menschen massgeblich beeinflussen. Zum Beispiel gilt generell: „je kleiner desto schädlicher (pro Kilogramm)“. Dies liegt daran, dass kleine Partikel tiefer in die Lungen eindringen als grössere Partikel. Nanopartikel gelangen sogar in die Alveolen und können Zellwände durchdringen. So gelangen sie direkt in den Blutkreislauf und das Nervensystem, werden im ganzen Körper verteilt und können mit vielen Geweben interagieren. Eine differenzierte Bewertung von Feinstaub ist deshalb Bedingung für eine realistischere ökologische Bewertung verschiedener Feinstaub-Quellen.

Das Wirkungsabschätzungsmodell innerhalb der LCA soll hinsichtlich der Feinstaub-Problematik verbessert werden, ohne das System unendlich kompliziert zu machen. Das soll erreicht werden durch die Bildung von multidimensionalen Feinstaub-Kategorien, die aufgrund von physikalischen und chemischen Eigenschaften (z.B. Grösse, Morphologie, chemische Zusammensetzung, Oberfläche etc.) in Relation zu gesundheitlichen Auswirkungen gesetzt werden. Die neu definierten Feinstaub-Kategorien können dann in bestehende Sachbilanzen (z.B. in derecoinvent Datenbank) und Bewertungsmethoden integriert werden.

Ziel

Das Hauptziel der Ressourcenpolitik Holz (<http://www.bafu.admin.ch/aktionsplan-holz>), in Abstimmung mit der Waldpolitik 2020 des Bundesrates, besteht in der nachhaltigen Holzbereitstellung und ressourceneffizienten Verwertung. Bis zum heutigen Zeitpunkt ist eine ökologische Bewertung von energetisch verwertetem Holz nur bedingt möglich, da die Grundlagen für diese Bewertung lückenhaft sind.

Das Ziel dieser Arbeit ist deshalb eine Erweiterung und Verbesserung der Ökobilanzierungsmethode für eine bessere Entscheidungsfindung bei Systemen, in denen gesundheitliche Schäden beim Menschen durch Feinstaub eine wichtige Rolle spielen.

Um dieses Ziel zu erreichen, wurden Zwischenziele zu den wichtigsten Arbeitsschritten formuliert:

- Bestehendes Wissen zu Partikeleigenschaften sammeln und geeignete chemische und/oder physikalische Eigenschaften selektionieren (Kapitel 2 und 3).
- Bestehendes Wissen zu Wirkungsabschätzung von Feinstaub zusammentragen (Kapitel 4 und 5).
- Ein Wirkungsabschätzungsmodell aufgrund der Partikeleigenschaften neu modellieren (Kapitel 6 und 7)
- Charakterisierungsfaktoren basierend auf chemischen und physikalischen Eigenschaften der Partikel berechnen (Kapitel 8 und 9)
- In einer Fallstudie soll aufgezeigt werden, wie sich die Resultate aus der Beurteilung des Feinstaubes verändern, wenn zur Beantwortung derselben Fragestellung einmal eine alte und einmal die neue Methodik verwendet wird (Kapitel 10).

Inhalt

Die Literaturrecherche zu relevanten chemischen und physikalischen Partikeleigenschaften hat ergeben, dass die Partikelmenge (Masse, Anzahlkonzentration, spezifische Oberfläche), Partikelgrösse, Wasserlöslichkeit, Morphologie/Form und die chemische Zusammensetzung der Partikel die Haupteinflussfaktoren bezüglich Toxizität am Menschen sind. Aufgrund der Literaturstudie wurden die Partikelmenge, chemische Zusammensetzung und Löslichkeit selektioniert. Diese drei Eigenschaften erscheinen zum heutigen Zeitpunkt geeignet, um in ein Wirkungsabschätzungsmodell implementiert zu werden. Diese Partikeleigenschaften eignen sich deshalb, weil sie klar definierbar und quantifizierbar sind (Voraussetzung der Lebenszyklus-Inventarmodellierung), einen direkten Zusammenhang zu schädlichen Effekten am Menschen haben und die jeweilige Eigenschaft für einen grossen Teil der Feinstäube in der Umwelt relevant ist.

Die Literaturrecherche zu Feinstaub-Ausbreitungsmodellen hat gezeigt, dass die am häufigsten gebrauchten Wirkungsabschätzungsmodelle (z.B. Ecoindicator 99, ReCiPe) sehr einfache Ausbreitungs- und Expositionsmodelle implementiert haben. Wichtige Prozesse sind nicht oder nur ungenügend berücksichtigt. Keines dieser bekannten Modelle berücksichtigt zum Beispiel die Zusammenballung einzelner Partikel (Koagulation). Gerade dieser Prozess ist aber sehr wichtig wenn Nanopartikel berücksichtigt werden sollen.

Die Literatursuche zu Exposition hat aufgezeigt, dass es sehr wichtig ist zu ermitteln, wie viele der eingeatmeten Partikel tatsächlich in der Lunge verbleiben und wie viele umgehend wieder ausgeatmet werden. Dies hängt sehr stark von der Grösse der Partikel ab. Aber auch das wird in den bekanntesten Modellen bei der Bewertung von Feinstaub nicht berücksichtigt.

Die Literaturarbeit zu Gesundheitseffekten, beruhend auf epidemiologischen Studien, hat ergeben, dass von den vorher genannten Effekten (Partikelmenge, chemische Zusammensetzung der Partikel und Löslichkeit) nur für die Partikelmenge eine gute Datenbasis vorhanden ist. Löslichkeit von Feinstäuben wird in epidemiologischen Studien nicht behandelt. Es gibt nur wenige epidemiologische Studien, welche Gesundheitseffekte mit der chemischen Zusammensetzung von Feinstäuben assoziieren.

Basierend auf allen Ergebnissen der Literaturstudien wurden folgende Anforderungen für die Modelle formuliert:

- Ausbreitungsmodell: Da die Bewertungsmethode den Schaden verschiedener Partikelgrössen unterscheidet (unter anderem Nanopartikel), wurde Koagulation als Elimination der Anzahlkonzentration im Modell integriert. Weiter wurden berücksichtigt: Ausbreitung innerhalb der geographischen Systemgrenze, Partikel-Aufenthaltszeit, Partikel Auswaschung durch Regen, Partikelsedimentation, Höhe über Boden an dem der Staub emittiert wird.
- Expositionsmodell: In Abhängigkeit der Partikelgrösse von 1 nm bis 10 µm ist die Menge an Partikeln ermittelt worden, welche einerseits eingeatmet wird, andererseits aber auch tatsächlich in der Lunge zurück gehalten wird. Weitere Determinanten des Expositionsmodells sind: Populationsdichte, Ort der Emission (Stadtzentrum, urban, ländlich).
- Das Effektmmodell beinhaltet die drei Effekte Partikelgrösse, chemische Substanz und Löslichkeit
 - 1) In diesem Modellierungsschritt wurden die Effekte von Partikelgrösse, chemische Zusammensetzung des Feinstaubes und dessen Löslichkeit auf die menschliche Gesundheit bewertet. Effekte verschiedener Partikelgrössen wurden anhand einer umfassenden Literaturarbeit zu epidemiologischen Studien berechnet. Die Resultate haben ergeben, dass Nanopartikel (Partikel < 100 nm) einen mehr als doppelt so grossen Effekt auf den Menschen haben im Vergleich zu PM₁₀. Der Effekt von PM_{2.5} ist etwa 1.6 Mal so gross wie derjenige von PM₁₀.
 - 2) Die Effekte verschiedener chemischer Substanzen wurden ebenfalls mit epidemiologischen Studien zu ermitteln versucht. Allerdings ist festgestellt worden, dass die Datenlage dazu nicht ausreicht. Es gibt zu einigen wichtigen Komponenten von Luftschadstoffen keine epidemiologischen Daten (z.B. Dioxine und Furane), zu vielen andern Substanzen ist die Unsicherheit in den Resultaten sehr hoch. Daher wurde für die Effekte chemischer Substanzen Toxikologiedaten des „New Jersey Department of Environmental Protection“ verwendet. Die

Datenbank liefert Toxikologiewerte von über 240 chemischen Substanzen zu Lungenkrebs und Herz-Lungenkrankheiten.

- 3) Löslichkeit wurde mittels theoretischen Überlegungen in das Modell eingebracht. Lösliche Partikel verlieren den Effekt der Grösse, da sich die Partikel beim Einatmen auflösen. Hingegen kommt die ganze potentielle Toxizität aller chemischen Substanzen des Staubes zum Vorschein. Unlösliche Partikel behalten die Wirkung der Grösse, andererseits ist die Wahrscheinlichkeit, dass die toxische Substanz des Partikels mit der Zelle interagieren kann, reduziert, denn die toxische Substanz könnte teilweise oder komplett umgeben sein von unlöslicher Materie.
- Modellierung des Schadens an menschlicher Gesundheit: Die gesamten Gesundheitseffekte aller chemischer Substanzen wurden in „Disability Adjusted Live Years“ (DALY) umgerechnet. Diese DALY sind für Lungenkrebs und Herz-Kreislaufkrankungen bekannt. Das Modell verteilt den totalen Schaden in DALY auf die ganze für Europäische Verhältnisse gültige durchschnittliche Umgebungsluft. Dies wird unter Berücksichtigung der Resultate des Ausbreitungsmodells, des Expositionsmodells und des Effektmodells gerechnet. Die Schadenssumme des ganzen Feinstaubes, welcher in Partikellöslichkeit, Partikelgrösse und Partikelchemie unterteilt wurde, ergibt die bekannte Anzahl DALY. Die Summe der Massen all dieser Luftkomponenten ergibt die Masse des gesamten Feinstaubes in der Luft innerhalb der geographischen Systemgrenze.

Fünf kleine Fallstudien wurden durchgeführt um zu testen, ob das Konzept des neuen Modells funktioniert. Mit den Fallstudien wird aufgezeigt welche Möglichkeiten das neue Modell bietet. Daneben bieten die Fallstudien eine Möglichkeit zur Validierung des Modells. Die Resultate aus dem neuen Wirkungsabschätzungsmodell wurden mit Resultaten von etablierten Wirkungsabschätzungsmodellen verglichen. Das neue Wirkungsabschätzungsmodell berechnet Gesamtschäden am Menschen für Feinstaub aus Verbrennungsprozessen etwa in der gleichen Grössenordnung wie 2 andere oft verwendete Bewertungsmethoden (ReCiPe, Ecoindicator 99). Wenig toxische Feinstäube dagegen haben in einer Fallstudie massiv geringere Schäden am Menschen ergeben (5 bis 6 Grössenordnungen kleiner). Daraus kann geschlossen werden, dass das neue Wirkungsabschätzungsmodell verschiedene Feinstäube differenziert bewertet. Die Resultate können einfach aufgeschlüsselt und dargestellt werden aufgrund der Partikelgrösse, der chemischen Zusammensetzung, der Löslichkeit, dem Ort und der Höhe über Boden der Emission sowie der Populationsdichte im betroffenen Gebiet.

Die wichtigsten Ergebnisse aus diesem Projekt sind:

- Das vorhandene Wissen zur Ausbreitung von Partikeln in der Atmosphäre in Abhängigkeit der Partikelgrösse und das Wissen zu toxischen Effekten von Partikeln bezüglich der Gesundheit des Menschen zum Zweck der Modellierung von Wirkungsabschätzungsmodellen wurde auf aktuellem Stand zusammengefasst und strukturiert.

- Bis anhin gibt es für die Bewertung von Partikeln in Ökobilanzen nur 1 Charakterisierungsfaktor, nämlich PM₁₀. Dies bedeutet, dass ein harmloser, grosser Partikel aus dem Ackerbau gleich bewertet wird wie ein hochgiftiger, nanoskaliger Partikel aus Dieselabgasen. Neu wurden 2781 verschiedene Charakterisierungsfaktoren berechnet.
- Die neue Modellierung der Gesundheitseffekte basiert auf chemischen und physikalischen Eigenschaften der Partikel (Grösse, Chemie, Löslichkeit). Daneben wurden weitere Parameter berücksichtigt, welche signifikanten Einfluss auf die Exposition der Menschen haben: 1. Höhe der Emission (Hochkamin in der Industrie, Hausdach oder mobile Quelle am Boden); 2. Der Ort der Emission (Stadtzentrum, urban oder ländlich Umgebung); 3. verschiedene Populationsdichten (Stadtzentrum, urban, ländlich).
- Basierend auf den Resultaten aus den Literaturstudien (Ausbreitung, Exposition und Effekte) wurden neue, verbesserte Modelle erfolgreich umgesetzt. Das Ausbreitungsmodell zum Beispiel ist das einzige den Autoren der Studie bekannte Modell für Ökobilanzanwendungen welches Koagulation von Nanopartikeln berücksichtigt.

Methodenkritik

Die Methode hat trotz starker Anpassungen von etablierten Wirkungsabschätzungsmodellen immer noch Mängel:

- Als Schwäche können die immer noch erheblichen Unsicherheiten von chemischen Substanzen bezüglich ihrer Toxizität erwähnt werden. Dies lässt sich erklären da die Daten zu Toxizität aus toxikologischen Studien stammen. Im Idealfall würde man nur epidemiologische Studien zur Berechnung der Wirkung am Menschen aufgrund der Chemie verschiedener Stäube benutzen. Solche Daten werden aber in den nächsten 10 – 20 Jahren vermutlich noch nicht in ausreichender Menge vorhanden sein.
- Eine wichtige Kategorie von Feinstäuben – die sekundären organischen Stäube – konnten nicht implementiert werden. In Zusammenarbeit mit Experten aus Atmosphärenchemie, Atmosphärenphysik und Klimamodellierung musste geschlossen werden, dass der Stand der Forschung heute noch nicht weit genug ist, um sekundäre organische Partikel in unser Modell zu implementieren.
- Die hohe Auflösung bezüglich Schäden am Menschen, welches das neue Schadensmodell bietet, erfordert einen massiv grösseren Aufwand in der Datenbeschaffung für Inventare. Bis jetzt wurde ganz einfach die Masse von PM₁₀ bestimmt. Neu müssen die Stäube unterschieden werden bezüglich Grösse, Chemie und Löslichkeit.
- Es gibt eine Reihe von Prozessen, welche in diesem Modell nicht berücksichtigt sind. Nukleation (Partikel welche aus Gasen entstehen) ist im Gegensatz zur Koagulation nicht implementiert.
- Ebenfalls nicht modelliert wurden Interaktionen von Partikel mit Wasserdampf bei erhöhter Feuchte, zum Beispiel in Wolken oder Nebel. Ähnlich wie bei der Bildung sekundärer Partikel werden diese Prozesse (chemische Umwandlung,

Agglomeration) heute noch zu wenig gut verstanden, um sie in ein Wirkungsabschätzungsmodell einzubauen.

Ausblick

Im Idealfall wird das neue Modell in eine etablierte Ökobilanzmethode eingebaut und ersetzt dort das bisherige Wirkungsabschätzungsmodell für Feinstaub. Dazu müssen aber Inventare erstellt werden, welche die Feinstäube aufgrund der genannten Merkmale unterscheiden können.

Executive summary

Polluted air with tiny dust particles is one of the greatest challenges in the air pollution control policy. Especially cities and traffic areas suffer from high levels of particulate matter. In these areas the yearly average values for particulate matter (PM) over the threshold values regulated by law. Epidemiological studies have consistently shown that PM lead to an increase in cardiac and respiratory diseases for humans. Airborne particles are composed of solid and liquid shares and come from various sources: combustion processes, mechanical abrasion of tires and road surfaces, degradation processes (e.g. from rock) etc. The combustion of wood products is often seen as problematic in terms of PM released to air, without ever having made an objective, comprehensive assessment of the advantages and disadvantages of this raw material.

The method of Life Cycle Assessment (LCA) is often used to optimize environmental impact of processes or products or to compare the effect of different processes or products with each other. LCA provides a framework for the identification and evaluation of the environmental impacts (including impacts on humans) over the entire life cycle (from raw materials extraction to disposal) of products or processes. For example, a LCA assesses potential damages to human health, to ecosystem and resources caused by the operation of a wood-burning system and compares these damages with those caused by the operation of an oil/gas combustion system, or the wood log combustion can be compared with the combustion of wood pellets. The prerequisite for this is that emissions with different effect can be distinguished in the LCA.

Today, LCA-considers particulate matter undifferentiated as PM₁₀ (particles size less than 10 µm). It is well known, however, that there are many important particle properties, such as size, water solubility, chemical toxicity, which have significant influence on the effects on human health. For example, it is generally accepted that "the smaller the particles, the more harmful (per kilogram) ". This is due to the fact that small particles penetrate deeper into the lung compared to larger particles. Nanoparticles even get to the alveoli and can even penetrate cell walls. So they get directly into the blood stream and will be distributed in the whole body and can interact with many tissues and organs. Therefore a differentiated evaluation of particulate matter (PM) is a prerequisite for more realistic environmental evaluation of different fine dust sources.

The impact assessment model within the LCA is to be improved in terms of problems due to PM without making the system infinitely complicated. This will be achieved through the formation of multi-dimensional particulate matter categories that are set on the basis of physical and chemical properties (e.g. size, morphology, chemical composition, surface, etc.) and related to human health effects. The PM categories newly defined can then be integrated into existing life cycle inventories (e.g. in the ecoinvent database) and impact assessment methods.

Objective

The main objective of the wood resource policy (www.bafu.admin.ch/aktionsplan-holz) in accordance with the forest policy 2020 is a sustainable wood supply and efficient use of wood. To date, a meaningful ecological assessment of wood energy is only possible to a limited extent, because the basis of the assessment is incomplete.

The objective of this work is therefore an extension and an improvement of the LCA method for better decision making in systems in which damage to human health caused by PM play an important role.

To achieve the objective, intermediate objectives have been formulated:

- The literature is to be reviewed to the existing knowledge of particle properties and appropriate chemical and / or physical properties shall be selected (Chapter 2 and 3).
- The literature is to be reviewed to the existing knowledge about impact assessment modelling of particulate matter (Chapter 4 and 5).
- A new impact assessment shall be modelled based on the particle properties and demands on impact assessment models defined before (Chapter 6 and 7),
- Characterisation factors have to be calculated based on chemical and physical properties of the particles (chapters 8 and 9).
- A case study has to have to prove concept and shall demonstrate how the results from the assessment of particulate matter change if for the same problem once an old and once the new methodology is used (Chapter 10).

Content

The literature search of relevant chemical and physical particle properties has shown that the amount of particles (measured as mass, number concentration, surface area), particle size, water solubility, morphology/shape, and chemical composition of the particles are the main factors for toxicity in humans. Based on the literature study, the particle size, chemical composition and solubility were selected as suitable properties.

These particle properties were designated as suitable because

- they are clearly definable and quantifiable (prerequisite for life cycle inventory modelling)
- they show a direct relationship to cause harmful effects on humans and
- the property is relevant for a large part of the PM released to ambient air.

The literature search on PM dispersion models has shown that the most commonly used impact assessment models (e.g. Ecoindicator 99, ReCiPe) relying on very simple dispersion and exposure models. Important processes are not or only insufficiently taken into account. None of these known models considered for example coagulation. In particular this process is very important when nanoparticles are taken into account.

The literature search on exposure showed that a good model should determine the amount of inhaled amount particles. Even more important, but mostly neglected, is to determine the amount of particles remaining in the lungs and how much is exhaled again.

This depends very much on the size of the particles. Again, the mass taken up in the lung is not taken into account in the most common used impact assessment models for PM.

The literature search on health effects based on epidemiological studies has shown that a good database is available for the amount of particles only considering the previously mentioned PM properties (particle size, chemical composition of the particles and solubility). Solubility of PM is not addressed in epidemiological studies. There are few epidemiological studies which associated health effects with the chemical composition of PM.

Based on all the results of the literature review the following requirements for the models have been formulated:

- Fate model: Since the impact assessment distinguishes the damage of various particle sizes (including nanoparticles), coagulation was integrated as main elimination of the particle number concentration in the model. It was further considered: dispersion of PM within the geographical system boundary, particle residence time, particle wet deposition by rain, particle sedimentation and finally the height above ground at which PM is emitted.
- Exposure model: Depending on the particle size from 1 nm to 10 microns, the amount of particles has been determined which on the one hand is inhaled, on the other hand, in fact retained in the lung. Other determinants of the exposure model are: population density, location of the release (city centre, urban, rural).
- The effect model is based on the three effects particle size, chemistry of PM and solubility of PM.
 - 1) The human health effects of particle size, chemical composition of PM and its solubility were evaluated. Effects of different particle sizes were calculated from a comprehensive literature review to epidemiological studies. The results have shown that nanoparticles (particles < 100 nm) have an effect on humans which is more than twice as large compared to PM₁₀. The effect of PM_{2.5} is about 1.6 times as large as that of PM₁₀.
 - 2) The effects of various chemical substances have been determined with epidemiological studies. However, it has been found that the data location is currently too small. For some important components of air pollutants (e.g. dioxins and furans) no epidemiological data at all were found. For many other substances the uncertainty in the results is simply too high. Therefore, toxicology data of the "New Jersey Department of Environmental Protection" was used for the effects of chemical substances. The database provides toxicology data for more than 240 chemical substances which relate to lung cancer and cardiopulmonary diseases.
 - 3) Solubility was incorporated into the model by means of theoretical considerations. Soluble particles lose the effect of the size as the particles dissolve by inhalation. On the other hand, the toxicity from all chemical substances of PM develops its full potential. Conversely,

insoluble particles retain the effect of the size, on the other hand the probability that the toxic substance of the particle can interact with the cell is reduced, because the toxic substance can be partially or completely surrounded by insoluble not toxic matter.

- Damage model: the total health effects of all chemical substances were converted in "disability adjusted live years" (DALY). This DALYs are known for lung cancer and cardiovascular diseases caused by PM. The model distributes the total damage expressed in DALYs over the whole PM of average ambient air in the European context. Here, the results of the fate model, the exposure model and the effect model are also included. The summed total of all PM fractions that were split in effects of size, chemical substance and solubility yields the known number of DALYs. The sum of the masses of all these components yields the mass of the entire PM in the air within the geographic system boundary.

Five small case studies were conducted to test whether the concept of the new model works. The case studies demonstrate the possibilities the new model offers. In addition, the case studies provide a way to validate the model. The results from the new impact assessment model were compared with results of established impact assessment models. For combustion processes the new method assesses the total damage to human health for PM in approximately the same order of magnitude as two other commonly used methods (ReCiPe, Ecoindicator 99). PM with low toxicity leads to results far below the results of other methods (5-6 orders of magnitude smaller). It can be concluded that the new impact assessment model successfully differentiates between PM fractions with high and low toxicity. The results can be easily broken down and presented reflecting particle size, chemical composition, solubility, the location and height above ground of the emission release and the population density in the affected area.

The main results from this project are:

- The existing knowledge on fate of particles in the atmosphere as a function of particle size and the knowledge on toxic effects of particles on human health considering the use in impact assessment modelling has been summarized structured.
- Up to now, there was only one characterization factor in LCA for the evaluation of particles. This means that a harmless, large particle from the agriculture land processing is valued the same as a highly toxic, nanoscaled diesel soot particle. The new impact assessment provides 2781 different characterization factors for the characterisation of PM.
- The new modelling of health effects is based on chemical and physical properties of the particles (size, chemistry, solubility). In addition, further parameters, which have significant influence on the exposure of human are taken into account: 1. Height above ground of the emission (high stack in the industry, low stack of house or mobile source on the ground); 2. The location of the emission (city centre, urban or rural area); 3. Different population densities (city centre, high and low population density).

- Based on the results from the literature studies (fate, exposure and effects) new, improved models have been successfully implemented. The fate model is to our knowledge the only model for LCA applications, in which coagulation of nanoparticles is considered.

Critique of methods

Despite strong adaptations of established impact assessment models there are still some shortcomings and limitations:

- There is still a considerable amount of uncertainty reflected in the toxicity of the chemical substances. This can be explained because the data on toxicity stem from toxicological studies. Ideally, one would use only epidemiological studies to calculate the effect on humans due to the chemistry of various PM components. Such data, however, will probably not yet be available in sufficient quantity in the next 10 - 20 years.
- One important category of fine dust - the secondary organic PM - could not be implemented. In collaboration with experts from atmospheric chemistry, atmospheric physics and climate modelling, we had to conclude that the state of research is not far enough today to implement secondary organic particles in our model.
- The high resolution of damage to human health due to the number of chemical species, sizes classes, solubility, etc. requires a much larger effort considering data collection in order to inventory PM. Until now, the mass was easily determined by measuring PM₁₀ mass. Now the PM₁₀ has to be distinguished in terms of size, chemistry, and solubility.
- There are a number of processes which are not considered in this model. For example, nucleation (particles formed from gases) in contrast to the coagulation is not implemented.
- In addition, we neglected interactions of particles with water vapour in the air, for example, in clouds or fog. Similar to the formation of secondary particles, such processes (chemical conversion, agglomeration) are not understood well enough in order to integrate them in an impact assessment model.

Outlook

Ideally, the new model is incorporated into an established impact assessment method, where it replaces the existing impact assessment model for PM. For that purpose, inventories would have to be created which can distinguish PM due to the mentioned characteristics.

Table of content

1	Glossary/Abbreviations	4
2	Introduction.....	6
2.1	The relevance of Particulate Matter.....	6
2.2	Project goals	9
2.3	A short introduction into LCA.....	10
2.4	Line of sight - The project approach.....	15
3	Literature screening on the relevance of particle properties	16
3.1	Mass/number	16
3.2	Number concentration/surface area	16
3.3	Particle size.....	16
3.4	Water solubility.....	17
3.5	Morphology	17
3.6	Chemical composition	18
3.7	Crystallinity.....	18
3.8	Reactive Oxygen Species (ROS).....	19
4	Selection of particle properties	20
4.1	Size.....	20
4.2	Amount (mass/number concentration).....	20
4.3	Number concentration/surface area	21
4.4	Water solubility	21
4.5	Morphology/shape.....	21
4.6	Chemical composition	21
4.7	Crystalline structures	21
4.8	ROS.....	21
5	Literature to fate and exposure	22
5.1	State of the art fate models in LCA.....	22
5.2	Overview over Exposure modelling in LCA.....	23
6	Literature review of human health effects.....	25
6.1	A short introduction into odds ratio and relative risk.....	25
6.2	Literature review of epidemiology studies on the association between exposure to particulate matter and human health outcomes	25
6.3	Chemical composition of PM.....	35
6.4	Evaluation of the Literature for PNC of UFP and precursor gases of secondary particles (SO _x , NO _x and CO).....	35

6.5	Inverse Variance method: Weighting studies for meta-analysis purpose.....	38
6.6	Evaluation of the Literature for the chemical composition of PM	40
6.7	PAH, Dioxin and other potentially toxic substances	42
6.8	Unit risk factor and reference concentration	42
7	Description of PM in ambient air based using size classes and chemical composition	43
7.1	Main components.....	43
7.2	Trace elements	44
7.3	Organic matter	44
7.4	Dioxins and dioxin-like compounds.....	44
8	Modelling Fate, Exposure, Effects and Damage	46
8.1	Modelling the background concentration.....	46
8.2	The Fate Model.....	47
8.3	The exposure model	53
8.4	The effect model	56
8.5	The damage model	61
9	The characterisation factor (CF)	63
9.1	Base model.....	63
10	Verification of the Model	64
10.1	Size and solubility	64
10.2	Population density.....	64
10.3	Stack height.....	65
10.4	Chemical substances	66
10.5	Allocation of damage to human health by chemical components of PM in ambient air	66
11	Case studies	69
11.1	Case study 1: Residential wood combustion of different types of wood	69
11.2	Case study 2: two wood-based household heating systems	71
11.3	Case study 3: Small-scale wood combustion with different stoves	72
11.4	Case study 4: Mineral dust from sawing natural stone	77
11.5	Case study 5: Heat from light fuel oil vs. heat from pellets	79
11.6	The new impact assessment versus the established ReCiPe and Ecoindicator 99 methods	89
12	Monetarisat ion of damage – valuation of a life year (VOLY).....	91
13	Perspective: New impact assessment and the ecological scarcity method ...	95
14	Project resume and outlook	98
14.1	Achievement of objectives.....	98

14.2 Limitations of the new impact assessment.....99

14.3 Follow up projects 100

14.4 Outlook.....100

15 Acknowledgment..... 102

16 Literature 103

17 Index of tables 114

18 Index of figures..... 116

19 Annex 118

1 Glossary/Abbreviations

CF:	Characterisation factor – a factor derived from a characterization model which is applied to convert an assigned life cycle inventory analysis result to the common unit of the category indicator
CHF:	Congestive heart failure
CI:	Confidence interval
COPD:	Chronic obstructive pulmonary disease
CPD:	Cardiopulmonary disease
CVD:	Cardiovascular disease
DALY:	The disability-adjusted life year is a measure of overall disease burden, expressed as the number of years lost due to ill-health, disability or early death
Damage model:	translates the human health effects into damage to human health
ecoinvent:	A life cycle inventory database
ED:	Cardiovascular emergency department
Effect model:	Links the dose to human health effects
EI99:	Ecoindicator 99, a methodology for Life Cycle Impact Assessment
Exposure model:	links the concentration from the fate model to a dose.
Fate model:	Links the emissions to a concentration
IHD:	Ischemic heart disease
IQR:	Interquartile range
LCA:	Life cycle assessment
LCI:	Life cycle inventory
LCIA:	Life cycle impact assessment
OR:	OR represents the odds that an outcome will occur given a particular exposure, compared to the odds of the outcome occurring in the absence of that exposure
PAH:	Polycyclic aromatic hydrocarbon
PM:	Particulate Matter
PM ₁₀ :	PM ₁₀ standard includes particles with a diameter of 10 micrometres or less
PM _{2.5} :	PM _{2.5} standard includes particles with a diameter of 2.5 micrometres or less
PNC:	Particle number concentration
RD:	Respiratory disease
ReCiPe:	A methodology for Life Cycle Impact Assessment

- RFC: The Reference concentration is an estimate of a continuous inhalation exposure to the human population (including sensitive subgroups) that is likely to be without an appreciable risk of deleterious effects
- ROS: Reactive oxygen species
- RR: Relative risk is the ratio of the probability of an event occurring in an exposed group to the probability of the event occurring in a comparison, non-exposed group
- UFP: Ultrafine particles are particulate matter of nanoscale size (less than 100 nanometres in diameter).
- URF: The unit risk factor expresses excess lifetime cancer risk estimated to result from continuous exposure to an agent

2 Introduction

Atmospheric particles or dust (particulate matter, PM) have been considered a major component of air pollution. According to the BAFU the contamination of ambient air with PM₁₀ is the most relevant challenge of air pollution control in Switzerland. The impact of airborne particles on human health is today seen as the probably most important environmental issue in Europe. Recent assessments showed an expected loss in life expectancy of about 9 months in the year 2000¹ due to exposure to (ambient) PM_{2.5} mass. Epidemiological studies in recent years gave a strong evidence for increased morbidity and mortality due to cardiovascular disease and lung cancer, even at relatively low PM burdens.

PM₁₀ is composed of solid and liquid components and stems from many different sources: combustion processes, mechanical abrasion of tyres and road, degradation of minerals, etc. Polycyclic aromatic hydrocarbons (PAH), dioxins/furans and other harmful substances (heavy metals) were found in the PM mixture analysing dust from wood combustion. The existence of such substances creates cautionary voices claiming for strong regulations or even demanding a ban of wood for the production of heat and electric power. The question arises on what data base such direct conclusions are done, because the understanding of the corresponding causal chains of various parameters describing PM exposure and health effects is still incomplete. Physical and chemical differences of particles need to be better defined and included into health effect models or assessment tools. Many research projects on PM exposure and health effects due to PM have been initiated in the last years, mainly in the United States (EPA², HEI³) but also in Europe (COST⁴, CAFE⁵). The project from the Federal Office for the Environment „Ressourcenpolitik Holz“ aims at providing sustainable wood products with regard to economic and social, but also ecological aspects. A resource efficient utilisation of wood should allow high economic added value and at the same time minimizing environmental impacts along the life cycle.

2.1 The relevance of Particulate Matter

Life cycle assessment (LCA) is the method to be used when environmental benefits have to be assessed along the life cycle of a product. LCA is a technique to assess environmental impacts associated with all the stages of a product's life from cradle-to-grave (i.e., from raw material extraction through materials processing, manufacture, distribution, use, repair and maintenance, and disposal or recycling). To conclude, damage on human health due to PM is worldwide recognised as an important issue that needs further investigation. LCA is a suited tool to investigate environmental impacts of products along the life cycle. Unfortunately, state of the art LCA contains some major drawbacks when assessing human health effects due to PM₁₀.

More than 4000 datasets from many different industrial sectors are stored in ecoinvent, the most comprehensive life cycle inventory database worldwide. Among these datasets about 98% contain particulate emissions. The datasets represent the following activity fields: Agriculture (297 datasets), chemicals (369), materials (715), processing (268), transport

(193), energy (1259), waste management (409), and infrastructure (481). Thus, particulate emissions occur in almost all industrial activity fields.

Figure 1 shows the damage due to PM as a share of the damage to total human health (due to PM, radiation, carcinogenic substances, etc.) summarised for all processes and categorised into the above mentioned activities. It reveals that there are huge differences between the 3 impact assessment methods Impact2002+⁶, ReCiPe⁷ and Ecoindicator 99⁸ (EI99) and the 8 activity fields. The share of PM on total human health depends on both, the characterisation of PM (primary PM, secondary PM from SO_x, etc.) and from the midpoint categories (e.g. respiratory effects, climate change, human toxicity, etc.) summarised in the endpoint category Total Human Health. The comparison is partly biased since the different methods do not exactly reflect damage through the same substances. Ecoindicator includes primary particles and secondary particles from NO_x, SO_x and ammonia. ReCiPe covers the same substances as EI99 plus it accounts for the damage due to photochemical ozone creation. Impact 2002+ accounts for PM_{2.5} only. All methods consider ozone layer depletion, radiation, respiratory effects, human toxicity and photochemical oxidation to account for Total damage to Human health. Climate change is not included in Impact 2002+, but it is included in EI99 and ReCiPe. This might explain a part of the big difference between Impact 2002+ and EI99 or ReCiPe.

However, regardless on the industrial activity category and the impact method, PM contributes significantly to the damage to human health. Damage due to PM is dominant for Impact 2002+ – up to 80%, but only 20% for ReCiPe.

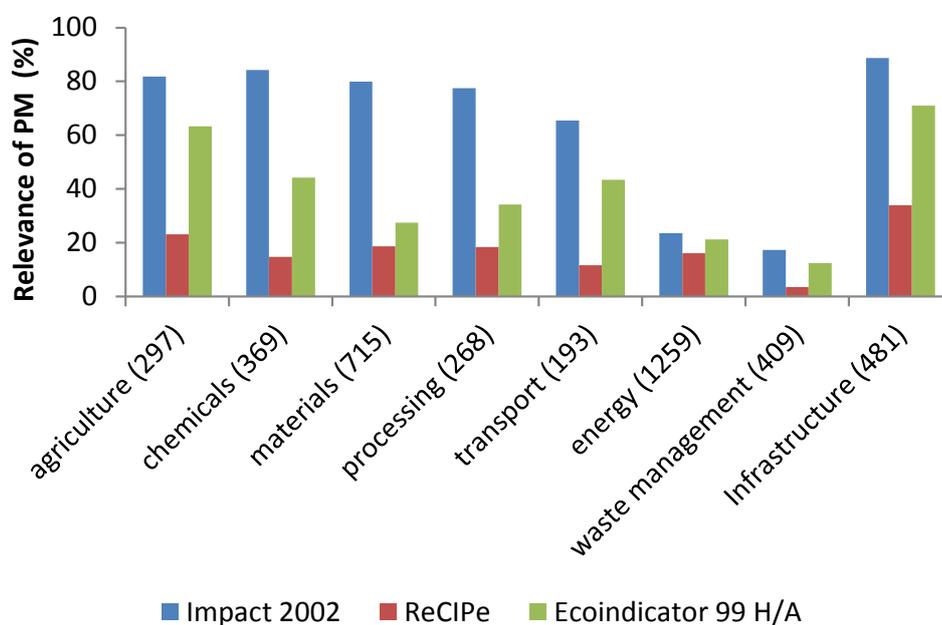


Figure 1. Damage due to PM as a share on total human health assessed with 3 different methods.

Shortcomings in LCA with respect to PM

Life cycle inventory

Size is one property of particles very often mentioned discussing the relevance of PM₁₀ emissions. PM is divided in 3 size classes in the ecoinvent database (PM>10 µm, 10 µm <PM >2.5 µm; PM<2.5 µm). No further characteristics are assigned to particles (chemical composition, solubility, morphology, surfaces characteristics etc.) in life cycle inventories. This is already a strong limitation regarding possible resolution of damage to human health due to different characteristics of particles.

Life cycle impact assessment

The models which associate damage of an emission require data from the life cycle inventory of a process. Each type of emissions gets a number that links the amount of emission to damage. The number is called Characterisation Factor (CF). A short glance at CFs for PM emissions from some impact assessment methods are provided in Table 1:

In addition to PM, a number of harmful substances are assigned separately in LCA. This characterisation is done using a unit risk of a chemical – neglecting all other physical and chemical properties, such as size, solubility, morphology, etc.

Impact assessment method	Characterisation Factor
CML	kg 1,4-DCB-Eq/kgPM
Particulates, < 2.5 µm	8.20E-01
Particulates, > 2.5 µm, and < 10µm	8.20E-01
Ecoindicator 99	DALY/kg PM
Particulates, < 2.5 µm	7.00E-04
Particulates, > 2.5 µm, and < 10µm	3.75E-04
ReCIpe	DALY/kg PM
Particulates, < 2.5 µm	2.60E-04
Particulates, > 2.5 µm, and < 10µm	2.60E-04
Particulates, < 10 µm	2.60E-04
Particulates, < 10 µm (mobile)	2.60E-04
Particulates, < 10 µm (stationary)	2.60E-04
Ecological scarcity 2006	UBP/kg PM
Particulates, < 2.5 µm	1.50E+05
Particulates, > 2.5 µm, and < 10µm	1.50E+05
Diesel particulates	1.70E+07
Impact2002+	Points/kg PM
Particulates, < 2.5 µm	9.86E-04

Table 1. Characterisation Factors for PM from 5 different impact assessment methods

Most impact assessment methods provide a selection of possible classifications for PM into different size categories or emission sites. However, the characterisation, expressed with the CF, is unchanged. There is 1 exception: Ecoindicator 99 provides different values

for Particles < 2.5 µm and Particles between 10 and 2.5 µm. Apart from this exception, each particle is assessed in all impact assessment methods with one CF only. Thus, state of the art LCA does not differentiate between a highly toxic, nano-sized particle from diesel exhaust and a hardly toxic particle from mechanical treatment of agricultural land or a stone pit.

Double counting

Another drawback of the assessment of particulate emissions in LCA is that the classification for many substances is ambiguous. Dioxins, PAH or heavy metals are separately assessed and have a CF. These substances are under ambient conditions always in a solid state and thus contribute to the PM mass. Since the characterisation of the impacts is based on long term epidemiological studies using PM₁₀ to calculate the damage, there is damage double counted due to such substances. The list of double counted substances encompasses all the substances that have a CF but are in a solid state under ambient conditions: Dioxins, furans, PAH, heavy metals, many organic substances with a low vapour pressure and high boiling point.

2.2 Project goals

An assessment of environmental burdens for processes in which particulate emissions might play a decisive role is disputable and provides results with very high uncertainties. The aim of this project is to elaborate a new impact assessment for PM by doing a reclassification of ambient particles into the air and by computing new CFs based on chemical and physical properties of particles.

The work should be accomplished in three steps:

- structuring the actual knowledge for fate of particles and the toxicology of particles with a comprehensive literature search
- new models shall be developed based on the results of the literature search
- the models shall be tested in a case study and the results will be compared with results using established impact assessment methods

A short introduction into LCA discloses the crucial steps for the modelling. This helps to understand the focus put to the relevant topics in the literature search.

2.3 A short introduction into LCA

Life Cycle Assessment (LCA) is an established but evolving tool primarily designed for accounting and assessing the potential environmental impacts caused by products, processes, or activities. It aspires to:

- identify and quantify the energy and materials used in all stages of a products lifetime and the associated wastes released to the environment,
- assess the impact of those energy and material uses on the environment,
- evaluate opportunities for environmental improvement.

The life cycle approach goes far beyond a single individual unit operation (like e.g., manufacturing) to encompass the whole 'cradle-to-grave'-system; it takes into consideration function, and a functional unit perspective. Both criteria are especially important when competing products are analysed to allow a comparative assertion. A set of ISO Standards (14'040⁹ et seqq.) describes the methodological framework for LCA. It is carried out in four distinct phases (Figure2).

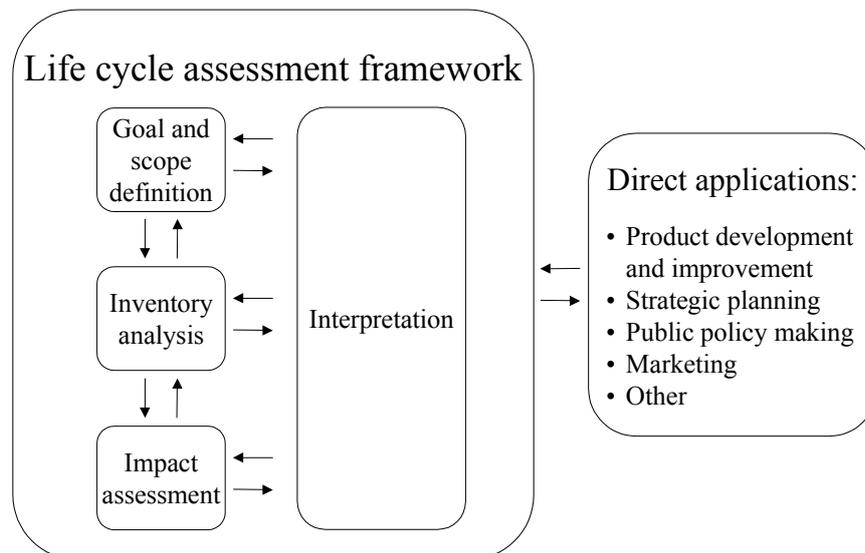


Figure 2. Phases of an LCA according to ISO 14040

Goal and scope definition

Goal and scope definition mainly sets out the context of the study, explains how and to whom the results are to be communicated and includes technical details that guide subsequent (functional unit, system boundaries, allocation methods, etc.). In this project this step will not be further discussed as it is not relevant.

LCI (life cycle inventory)

LCA is concerned with the whole life cycle of products "from cradle to grave". For the whole life cycle, the material and energy flows are put together to an inventory considering the scope and the goal of the study. The life cycle of a product is split into different modules (datasets) such as extraction of the raw materials, production or end of life. Thus the material and energy flows can be assigned to these modules which can be analysed in the impact assessment. LCI analysis involves creating an inventory of flows from and to

nature for a product system. Inventory flows include inputs of water, energy, and raw materials, released emissions to air, land, and water and waste flows to incinerations or landfills.

Life cycle impact assessment (LCIA)

According to the standards of the ISO 14'040 series, the impact assessment phase of LCA is aimed at evaluating the significance of potential environmental impacts using the results of the life cycle inventory analysis. In general this process involves associating inventory data with specific environmental impacts and attempting to understand those impacts. Classical LCIA consists of the following elements:

- Definition of impact categories to be taken into consideration.
- Classification of inventory categories to impact categories.
- Calculation of the effect scores per impact category (characterization) / Discussion of non-quantified relevant environmental aspects.
- Normalization of effect scores.
- Grouping / Weighting of normalized effects.
- Data quality analysis (e.g. by sensitivity analysis).
- While the first three steps are mandatory, the others can be done voluntarily.
- There are different methodological frameworks for LCIA. Figure 3 shows the framework from the method Ecoindicator 99.

Interpretation

Life Cycle Interpretation is a systematic technique to identify, quantify, check, and evaluate information from the results of the life cycle inventory and/or the life cycle impact assessment. The results from the inventory analysis and impact assessment are summarized during the interpretation phase. The outcome of the interpretation phase is a set of conclusions and recommendations for the study. According to ISO 14040, the interpretation should include:

- identification of significant issues based on the results of the LCI and LCIA phases of an LCA;
- evaluation of the study considering completeness, sensitivity and consistency checks; and
- conclusions, limitations and recommendations.

A key purpose of performing life cycle interpretation is to determine the level of confidence in the final results and communicate them in a fair, complete, and accurate manner.

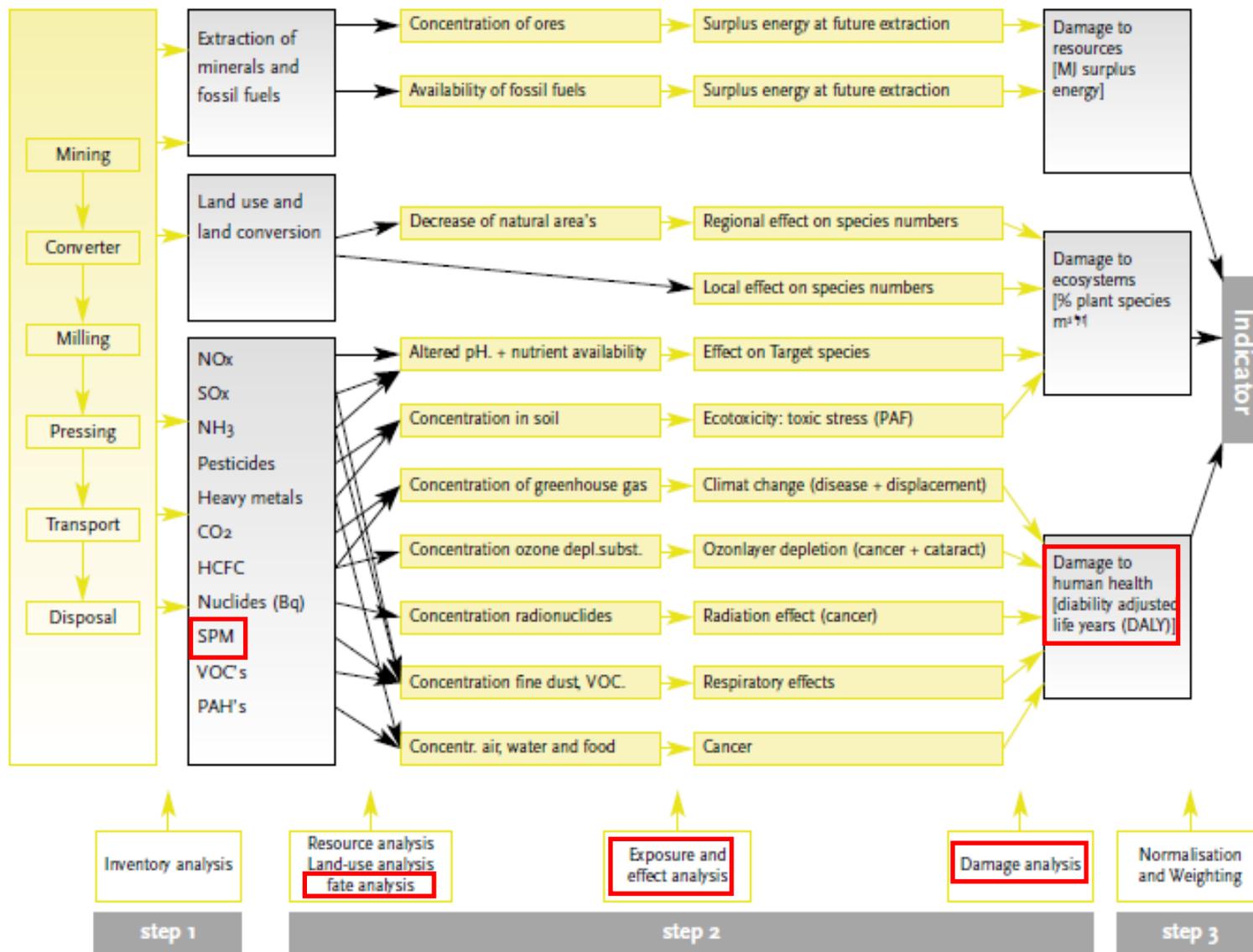


Figure 3. Framework for the impact assessment method Ecoindicator 99.

A closer look at life cycle impact assessment modelling

The impact assessment model associates inventory data with specific environmental impacts. For it CFs for all kind of emissions are calculated. A CF, e.g. for PM, is calculated using a fate model, an exposure model, an effect model and a damage model (Figure 3 and 4). With respect to the production of a good, PM might be released during several stages in a products life cycle. The emissions are listed in the LCI. In the fate analysis the emission is dispersed in the atmosphere and removal processes, dry and wet deposition, are considered. This leads to an exposure concentration. Exposure analysis determines the amount of emission taken up in the respiratory system. Exposure mainly depends on the exposure concentration and the Particle mass of inhaled particles that remains in the respiratory system after exhaling. Based on the mass taken up in the body and using any kind of data about toxicity (e.g. data from epidemiological studies, human intervention studies, animal studies, cell toxicological studies etc.) the effects on human health can be computed. This is called the effect analysis. The results are expressed as a number of cases, for example number of lung cancer cases, number of cardiovascular disease cases, etc. The effects have to be converted into a damage which is expressed as "Disability Adjusted Life Years (DALY). Finally, the CF is the result of a combination of all models.

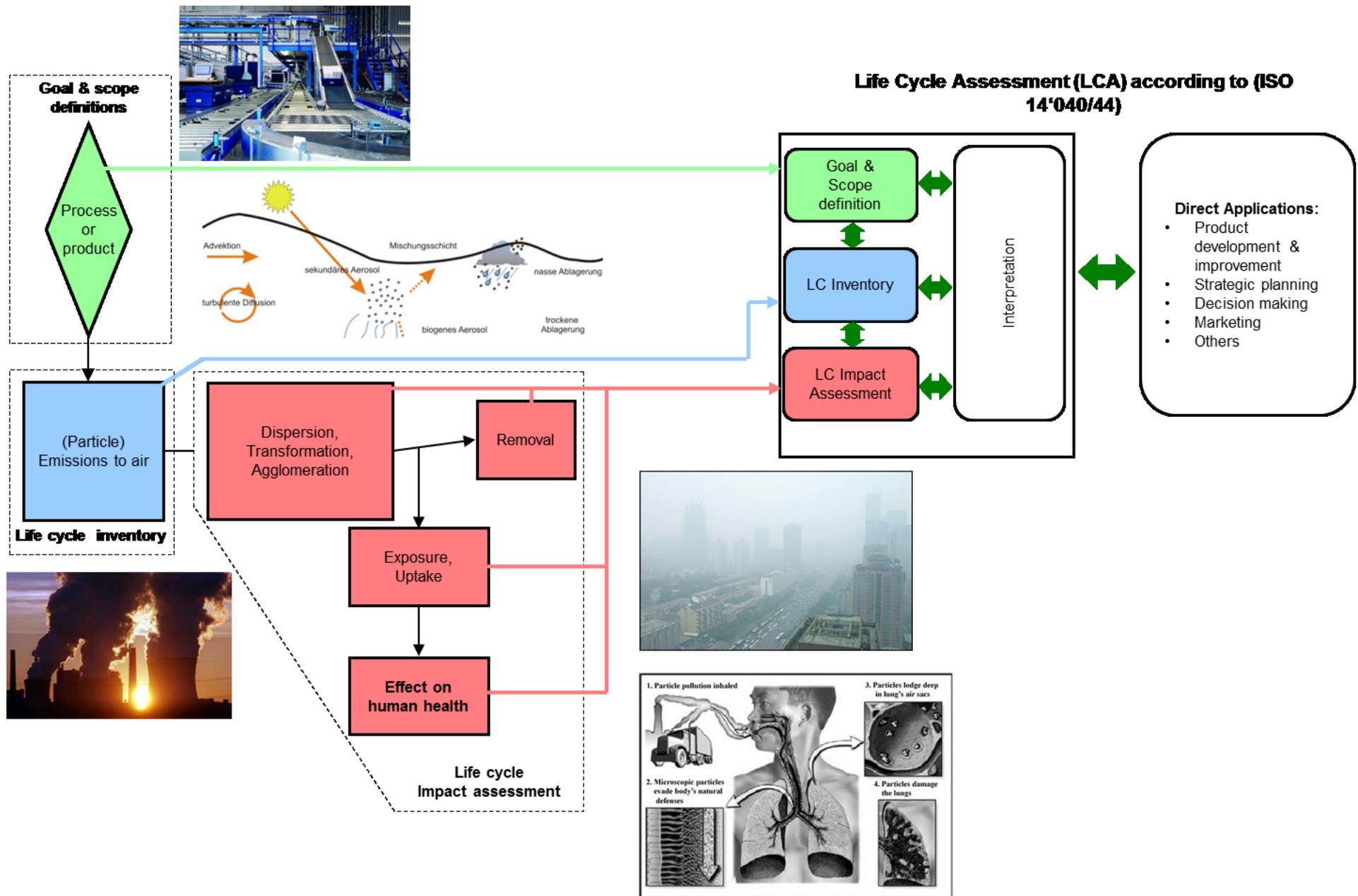


Figure 4. Steps required for the calculation of CF for PM emissions.
Human health damage characterisation factors for particulate matter emissions

2.4 Line of sight - The project approach

The project started by conducting a broadly based literature search that provides an overview over all relevant topics that have to be considered. This includes possible chemical and physical properties (size, chemical composition, particle shape etc.; chapter 2) suited to characterise PM and key issues with regard to the fate and the exposure model (chapter 4).

Suited properties were selected with advice from an expert group based on this first literature survey. Chapter 3 introduces the selected properties and gives reasons why certain properties were selected while others were discarded. According to the selection of properties a detailed literature study was then conducted. Thereby focus was put on direct links between effects as disease to real world conditions. Thus, the literature search encompasses exclusively epidemiological studies. These kinds of studies relate diseases such as lung cancer or CPD with ambient air concentration of PM (chapter 5).

A prerequisite for the selection of chemical and physical parameters to characterise damage to human health is that the parameter is measured in ambient air. Thus, a characterisation of European ambient air had to be achieved accounting on one hand on all substances with high shares on ambient air composition, but then also highly toxic substances found in extremely low concentrations (chapter 6).

The impact to human health due to PM emissions depends on the emission profile, the fate of PM in the atmosphere, the exposition of the population to the emission (intake and uptake fraction), the human health effects resulting from the exposition and the damage resulting in DALY. The CF is the number resulting from the entire impact assessment model and directly links the emissions to the impact. Chapter 7 provides all models derived for the new impact assessment model.

The model for the impact assessment can be written as:

$CF (DALY) = f (\text{Emissions; Fate; Exposure, HH-Effect, Damage})$, whereas the CF is a number that directly links an emissions from derived in life cycle inventories with damages resulting from the impact assessment models (Chapter 8).

Verification of the impact assessment model was derived by analysing the CF and conducting case studies. Analysis of the CF provides important information about whether the models work accurately (chapter 9). The case studies serve on hand as prove of concept for the impact assessment, on the other hand they provide information about the validity of the models by comparing the achieved results with results derived using other impact assessment models (chapter 10).

3 Literature screening on the relevance of particle properties

A first screening of literature revealed the most important chemical and physical particle characteristics. The characteristics most frequently mentioned in the literature are:

- amount (mass/number concentration)
- number concentration/surface area
- particle size
- water solubility
- morphology/shape
- chemical composition

3.1 Mass/number

It is obvious that the amount, either mass (in kg) or number (particle number concentration), of a release of particles to the environment are crucial elements on the LCI level in order to assess such a release in an appropriate way in the subsequent LCIA step. The framework for the calculation of LCA characterization factors allows for the consideration of nonlinear low-dose (mass or number) response curves. However, the availability of required data is limited in practice. Thus, assumption of a default linear low-dose-response relationship remains pragmatically necessary¹⁰. Consequently the amount is linear-causal related to human health effects.

3.2 Number concentration/surface area

Particle number concentration (PNC) is causally linked to the surface area. Once the distribution of and measure is identified it can be transformed easily into the other one (assuming spherical particles).

3.3 Particle size

Particle size is the only determinant considered for PM in LCA today. The size of a particle determines the penetration depth into the respiratory system (5). The deeper the particle penetrates, the higher gets the potential damage due to longer residence time in the respiratory system. Worst case is that a high amount of particles remain in the alveoli what then could end in silicosis. In vivo experiments show that small particles (constant mass concentration) produce higher acute inflammatory response than bigger particles^{11, 12}. Ultrafine particles (UFP, PM<0.1 µm) cross cell membranes, thus enter the blood stream and potentially can then be transported to any organ in the body¹³. Furthermore, it is known that UFP also are taken up by free nerve endings from the olfactory bulb reach this way the brain bypassing the blood-brain barrier¹⁴.

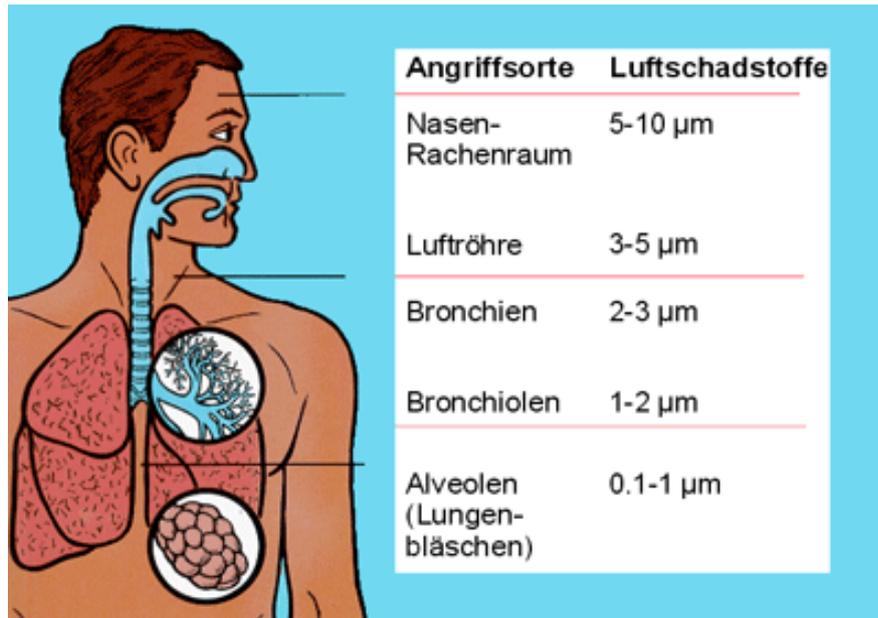


Figure 5. Penetration depth of inhaled particles depending on particle size.¹⁵

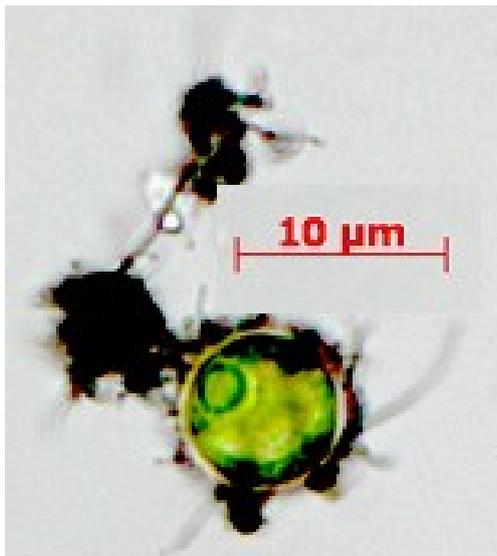
3.4 Water solubility

Depending on the solubility of a particle the interaction of particles with human tissues or organs in the body varies. Fast solving particles irritate the eyes, mucous membranes, or lead to inflammation of upper airways. Less fast solving particles affect cilia what curbs mucous removal. Poorly soluble particles might be solved not before reaching the alveoli. The consequences are cough, COPD, inflammation of bronchioles or alveoli.

Another aspect of solubility is that a particle is rather a mixture of different chemicals substances rather than a pure chemical. Toxic substances may attach to the surface or are partly or fully encapsulated in the matrix of a particle. A soluble particle releases all substances attached or encapsulated during inhalation. Consequently, toxic substances might develop their full hazard potential. For a non-soluble particle the hazardous substances remain within or attached to the particle and thus have reduced or even no impact of human health.

3.5 Morphology

The reactivity of a particle with its environment is mainly depending on the morphology or shape of the particle. Fibre shaped particles (e.g. asbestos fibres) are more reactive than spherical particles. Particles with reactive surfaces might attach to cells and completely inhibit signalling of the cell with its environment (Figure 6). The morphology of a particle also determines the ability to act as a transporter for toxic substances to penetrate into the body. Thus, morphology is an ambiguous term as it implies several important characteristics of a particle: e.g. shape, surface characteristics, transports capacity, etc.



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Figure 6. An algae cell surrounded by carbon black.

3.6 Chemical composition

PM is an inhomogeneous mixture of substances from different sources. It is composed from sea salts (evaporation of sea water), mineral dust (dispersed dust from soil), abrasion of tyres and on roads, abrasion from train, break wear, soot from incomplete combustion of wood or fossils, etc.

The chemical composition of particles decisively influences the potential damage to human health. Toxic particles might lead to significant damage on components of cells (e.g. mitochondria, cell membranes, genetic material). Heavy metals for instance accumulate in vital organs (nerves, brain, liver, kidneys, etc.). Heavy metals might replace essential minerals without taking over their functions. The list of potential disease is almost infinite: insomnia, fatigue, dizziness, anaemia, impotence, high blood pressure, osteoporosis, multiple sclerosis, etc.

3.7 Crystallinity

Crystalline structures have a potential to introduce fibrosis in the lungs. Freshly fractured quartz proved to have an increased potential to induce a fibrotic reaction in lung tissue. The presence of radicals on the fracture surface is the primary determinant in terms of toxicity. Hnizdo and Sluis-Cremer¹⁶ concluded that the risk to develop silicosis increase exponentially with an increase in cumulative dose of silica dust. SiOH groups on the surface of crystalline silica are capable to form hydrogen bonds with membrane components. The hydrogen bond formation leads to membrane damage and therefore disruption of cellular integrity¹⁷.

3.8 Reactive Oxygen Species (ROS)

ROS are chemically reactive molecules containing oxygen. ROS form as a natural by-product of the normal metabolism of oxygen and have important roles in cell signalling and homeostasis.

4 Selection of particle properties

The Expert group decided in a meeting about the final selection of the particle properties to be chosen for this project. Suitable physical and chemical properties had to meet the following restrictions:

- The property has to be clearly definable – overlap with other properties in the meaning should be avoided.
- General properties, valid, and relevant for most ambient particles should be chosen.
- The property must be quantifiable (with respect to the LCI).
- It is prerequisite that the property can be linked to human health effects. The best possible way in doing this is via results of epidemiological studies as these studies directly link the emissions with damage. Uncertainty in the damage increases strongly when the impacted assessment bases on data not representing the living human system. Thus, the second best would be to count of human intervention studies (humans exposed to particles in a lab), followed by animal studies and cell toxicological studies.
- The amount of selected properties should be limited so that the final set of CF is limited to a reasonable number.

4.1 Size

Particle size is considered to be the most relevant property for PM and it is easy to measure in terms of mass or number concentration. Particle mass for PM₁₀ is generally used in epidemiologic studies. Size will be subdivided into the following categories:

- Ultrafine particles (UFP <100 nm)
- Fine particles (100 nm - 2.5 µm)
- Coarse particles (2.5 µm – 10 µm)

Particles > 10µm will be neglected due to a low assumed impact on human health. This assumption is based on knowledge derived from the fate model (see paragraph 7.2) and the exposure model (see paragraph 7.3).

4.2 Amount (mass/number concentration)

Harrison et al.¹⁸ found that measuring the size distribution by particle count the number concentration has modes below 100 nm with the dominant mode being about 30 nm. However, mass modes peak at about 0.5 µm or higher and particles which contribute the most to particle number have little influence on mass concentration. The fact that ultrafine particles dominate the total particle count means that particle count measurements provide a better indication of human exposure to the UFPs. Vice versa, fine and coarse particles dominate the total particle mass measurement and this means that particle mass provides a better measurement of human exposure, whereas the number concentration of large particles contributes very little to the total number of particles measured. Hence, the amount for the three categories of size will be determined with the following measure:

- UFP (<100 nm): measuring PNC.
- Fine particles (100 nm - 2.5 µm): measuring particle mass.
- Coarse particles (2.5 µm – 10 µm): measuring particle mass.

4.3 Number concentration/surface area

A short glance at epidemiologic data reveals that PNC is related to damage on human health in a couple of studies, whereas there is not a large body of studies that related the effects of surface area on human health. Thus, number concentration will be used rather than surface area to measure UFPs.

4.4 Water solubility

The high relevance of water solubility deserves special attention. Although it will not be possible to directly link solubility with damages to human health, this topic will be accounted for by combining effects of size and effects of the chemical composition.

4.5 Morphology/shape

Morphology is an ambiguous term which implies several important characteristics of a particle: e.g. shape, surface characteristic, transport capacity, etc. However, due to the fact that these characteristics are not well described by this one term, morphology is discarded as a suitable property.

4.6 Chemical composition

No doubt chemical composition is a crucial property for the assessment of damage to human health. There is some literature linking chemical substances to human health effects in epidemiologic studies. However, there is not sufficient literature available to decipher the toxicity of the most relevant chemical components in an adequate way.

Thus, toxicology indicators are used from a database. It is important to use only 1 database for all substances rather than several different databases. Using 1 database only provides the advantage that it can be assumed that the consistency for the toxicology data is higher compared to using data from different data bases.

4.7 Crystalline structures

Crystalline structure was discarded as a property. It was regarded as too specific with only small relevance regarding the bulk of PM.

4.8 ROS

ROS has been considered as an important property, but there is not enough evidence in the prevailing literature, in particular from epidemiologic studies. ROS could be an important property for future improvements of PM assessment in LCA (time horizon: 10-20 years until first epidemiologic long term studies provide results).

5 Literature to fate and exposure

The literature survey for fate, exposure and effects focusses on one hand on the models used nowadays in LCA (fate and exposure) and on data availability for the effects of size and chemical composition (chapter 5).

5.1 State of the art fate models in LCA

Fate is the prediction of chemical and physical behaviour of a substance in the environment. The fate analysis aims at deriving fate factors for substances which are emitted to air and lead to an exposure by inhalation. The fate (distribution, dilution, deposition, etc.) of emissions causing respiratory effects are reflected in only one number: the fate factor. The fate factors transform emissions into external exposure (linking an emission to a concentration) that cause respiratory effects under ambient conditions. Some fate models have been analysed and the most relevant parameters and processes are summarised in Table 2.

Author	Hofstetter	Humbert	Rosenbaum	van Zelm	COSMO-ART	Austal
used in IA method:	Ecoindicator 99	-	Usetox	ReCIpe		
Model behind:	manual calc.	UFPM	Usetox	Eutrend	COSMO-ART	Austal
Dry deposition		x		x	x	x
Wet deposition	x	x		x	x	
Residence time	x					
Dispersion	x	x		x	x	x
Advection				x	x	x
chemical conversion of secondary organics						
Coagulation					x	
Dispersion	mean Europe	Distance to source variable	Europe/Global	50x50 km	variable	small scale
Particle characteristics	3 sizes	size, surface, number distributed	no PM	PM10	size distribution	

Table 2. Overview over main parameters and processes summarised for 6 fate models, partly applied in LCA impact assessment methods.

The most important processes of a fate model include deposition, dispersion and dilution and the residence time of particles in the atmosphere. Deposition or removal of particles is split into dry deposition (gravimetric settling of particles) and wash out (rain out). A third type of removal is coagulation. This removal process significantly reduces the number concentration whereas the mass remains unchanged. Coagulation is highly relevant for UFPs¹⁹. Removal processes influence residence time and thus also determine dispersion and dilution in the atmosphere. Figure 7 illustrates the relation between residence time and predominant removal process for particles in the lower 1.5 km of the atmosphere as a function of particle radius.

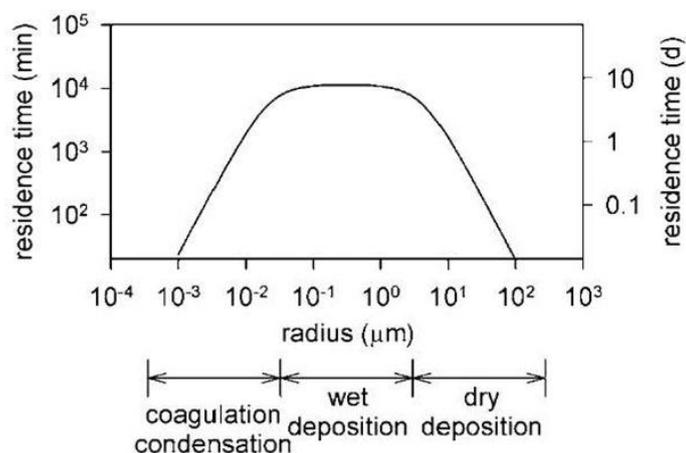


Figure 7. Residence time, particle radius and removal processes.

None of the fate models applied in LCA contains coagulation processes and none of these models include chemical reactions of secondary organics. The model developed by Hofstetter²⁰ is used in Ecoindicator 99⁸. This fate model is a very simple tool, that monodisperses an emission over a given area with a fixed mixing height. Wet deposition is the only removal process regarded. Austal²¹ does not consider wet deposition and coagulation processes. Furthermore, it is designed for dispersion on very small scale and focuses on boundary layers. Eutrend²² and COSMO-ART²³ are two very sophisticated models for air pollution dispersion modelling. The calculation of the results of fate factors for the present purpose would take weeks on a super computer. Thus, it is discarded for practical and economic reasons. USEtox is a model applicable for chemicals only. Particles and metals can't be modelled. The UFPM of Humbert²⁴ is a tool containing all relevant processes except coagulation. It does not address to a single particle size, but rather it accounts for particle size distribution.

The fate model to be developed should fulfil the requirements arising from the determined measures selected in chapter 3. Thus, a fate model should include, dispersion/dilution, residence time of particles in the atmosphere, gravimetric settling and rainout. With respect to UFPs coagulation should be implemented too, since we know that small particles have high effects on human health and they coagulate very fast. A particle size range of 1 nm to 10 μm should be covered. The model should represent European conditions.

5.2 Overview over Exposure modelling in LCA

Exposure assessment is the process of estimating or measuring the magnitude and duration of exposure to an agent, along with the number and characteristics of the population exposed. With exposure analysis the concentration derived in the fate model is linked to concentration response curves.

The damage assessment in Ecoindicator 99 (based entirely on Hofstetter²⁰ for PM) does not include an exposure analysis. Other methods^{22, 25} handle exposure by calculating the fraction of PM inhaled due to the exposition (intake fraction). The UFPM tool uses an advanced approach which allows assessing also an uptake fraction beside the intake fraction. The uptake fraction of PM is the fraction of deposited particles in the respiratory system. A summary of the processes used in the different methods is given in Table 3.

Exposure						
Author	Hofstetter	Humbert	Rosenbaum	van Zelm	COSMO-ART	Austal
used in IA method	Ecoindicator 99		Usetox			
Is exposure assessed?		x	x	x	x	
Intake fraction		x	x	x	x	
Breathing rate		x	x	x	x	
Degree of regionalization (population density)		6 types: urban, rural, remote, ...			variable according to the grid	
Uptake fraction		x				
Deposition fraction		x				

Table 3. Parameters and processes summarised for the same models as used for fate analysis. x = included in the model.

Figure 8 shows total average particle deposition and particle deposition in the alveolar, the tracheobronchial and the nasopharyngeal compartment²⁶⁻²⁸. Particles with diameter between 0.1 and 1 μm are mainly exhaled and not deposited in the respiratory system.

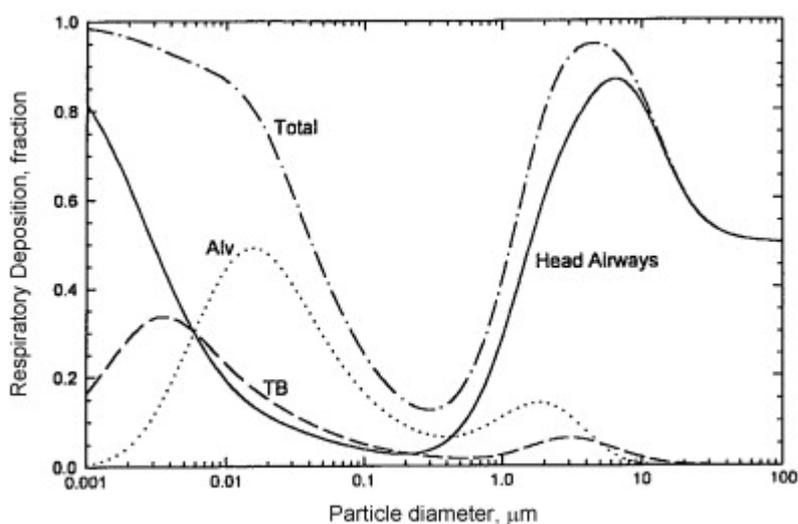


Figure 8. Lung deposition model.²⁶⁻²⁸

Neglecting this fact might lead to a wrong apportionment of PM to human health effects or at least to an increase of uncertainty. Thus, the exposure model should address a) the uptake fraction in the respiratory system, b) the intake fraction which depends itself on the time a person is exposed to ambient air and the breathing volume and c) the population uptake (total uptake of particles for the entire population exposed to the emission).

6 Literature review of human health effects

Existing LCA methods define PM in very general terms (i.e., total mass of PM). However, this evaluation of PM neglects the highly complex nature and variable health impacts of PM. This literature search aims to develop a more refined description of PM in LCAs that better reflects the contribution of PM to adverse effects than does mass alone. The purpose of the literature review described herein is to identify characteristics of PM that are associated with human health effects in epidemiology studies and that can be incorporated into the LCA methodology.

6.1 A short introduction into odds ratio and relative risk

The relation between particle concentration and health effect is described with relative risks (RR) or odds ratios (OR)²⁹. In statistics and mathematical epidemiology, RR is the risk of an event (or of developing a disease) relative to exposure. Relative risk is a ratio of the probability of the event occurring in the exposed group versus a non-exposed (less exposed) group. A RR of 1 means there is no difference in risk between the two groups. An RR of > 1 means the event is more likely to occur in the experimental group than in the control group. As long as the OR is around 1, there is more or less no difference between of RR and OR. When OR is more than 2.5 or less than 0.5 or probability of outcome in the unexposed group is more than 0.1 (10%), a correction should be applied^{30, 31}. In this study, values of OR higher than 1.1 and smaller 0.97 were discarded. Thus, in order to simplify the reading of this report all estimates of association with RR and OR are presented as RR only but it is referred to both, RR and OR.

Mean values of RR can be analysed with paired t-test in order to get an answer to the risk differences between the two compared substances. Paired sample t-tests analysis over the 95% CI (of each data-point provided in the literature studies) provides insight into the explained variance of the compared substances. The hypothesis is that the CI for each size class is smaller than CI for PM₁₀. This would support that the variance for the health outcomes is smaller when PM is split in different size classes.

Power analysis can be used to calculate the minimum sample size required so that it is reasonably likely to detect an effect of a given size^{32, 33}. Required sample size was calculated for a better interpretation of the analysed results from literature search. The sample size calculation was performed with an online power analysis tool³⁴ using a Power of 0.8 and a significance level α of 0.05.

6.2 Literature review of epidemiology studies on the association between exposure to particulate matter and human health outcomes

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Background

Airborne particles can come from a variety of sources and contain variable chemical constituents. Some particles are formed by natural processes, such as volcanoes, erosion, sea spray, and forest fires, while other are formed by anthropogenic processes, such as

industrial- and motor vehicle-related combustion, road-related wear, and mining. In general, larger particles (those greater than 2.5 µm) are formed by mechanical processes, while those less than 2.5 µm are formed by combustion processes. The chemical composition of particles is highly influenced by the source: for combustion-related particles, factors such as temperature of combustion, fuel type, and presence of oxygen or other gases can also have a large impact on PM composition. These differences can often be observed at a regional level, such as the greater sulphate-composition of PM in regions that burn coal for electricity production (which contains sulphur) versus regions that do not³⁵.

Most countries maintain air monitoring networks, and studies based on the resulting data are the most common basis for epidemiology studies on the health effects of PM. Data from these monitoring stations can be used to evaluate the relationship between community-level exposure to ambient particles and health outcomes (i.e., morbidity or mortality from various causes). Respiratory and cardiovascular outcomes are the most commonly assessed, although studies have also considered other related specific outcomes such as diabetes and congenital heart disease.

The data on particle characteristics is usually not very detailed and most often includes some combination of PM_{2.5}, PM₁₀, sulphate, and NO₂. Other descriptors that are less commonly found include particle number (ultrafine particles), metal components of PM, local traffic intensity, and EC/OC. Measures of association are usually reported per 10 µg/m³ or interquartile range increase in pollutant concentration. As the exposure data are taken from regional monitoring stations, the measurements are not representative of an individual's exposure.

Particle size is an important descriptor for understanding where in the human respiratory system the particles will deposit: as a general rule, smaller particles penetrate to deeper regions of the lungs. Initial studies on the health effects of particulate matter focused on mass of the particles, including either all particles (often termed total suspended particulate or TSP) or PM₁₀ (all particles with an aerodynamic diameter less than 10 µm). More recently, studies have considered both PM₁₀ and PM_{2.5}, with the latter corresponding more directly to combustion-related processes. UFPs are a dominant source of particles in terms of PNC, yet are negligible in terms of mass. Very few epidemiology studies have measured the effect of UFPs on health; however, the numbers of studies on this topic are increasing. In addition to size, chemical composition is of importance when understanding the toxicity of particles. Some studies consider the composition of particles in addition to mass; however this is not common, in part due the cost and labour involved in such analyses.

Methods

A literature review was conducted with the aims of identifying particle characteristics in addition to total mass that can be used to distinguish particle health effects. The literature on particle health effects is vast, and therefore a systematic review was not possible. Rather, priority was given to studies that:

- Were papers or reports of consensus opinions.
- Have had a significant impact on the understanding of human health effects or particles.
- Included a detailed analysis of particle composition or properties.
- Addressed specific sources of particles (i.e., source apportionment studies or studies of acute events [volcanoes, forest fires]).

Toxicology studies were not included. Although such studies play an important role in identifying mode of action and pre-clinical effects, limited efforts were focused on the most human-relevant data.

Results

Mass of PM_{2.5} and PM₁₀:

Mass-based measurements of particles are the most commonly-used measures in epidemiology studies on the effect of particulate matter on health. These measures are most easily and commonly collected, providing a large dataset for conducting such studies. For example, a quick search in PubMed using the search terms 'epidemiology', 'particulate', and 'matter' yields over 7000 hits. A review of all of the literature in this field is not feasible, but targeted reviews of existing health assessments of PM are informative.

The United States Environmental Protection Agency (US EPA) has undertaken perhaps the most thorough assessment of the health effects of particulate matter in their Integrated Scientific Assessment for Particulate Matter³⁶. The purpose of this assessment is to support US air quality regulations. This document, of which the most recent 2009 version numbers over 2000 pages, evaluates the existing knowledge available to estimate the effect of PM on human and environmental health, and includes studies on toxicology, epidemiology, controlled human exposure, atmospheric science, and other disciplines. This document provides a detailed analysis of the effects of PM_{2.5} and PM₁₀ in particular, however is primarily focused on data relevant to the United States. The sections on epidemiology studies include reviews of hundreds of epidemiology studies, and the conclusions based on these reviews are summarized below.

The EPA ISA states the following regarding the effects of long and short term exposure to PM_{2.5} and PM₁₀. For more details on any individual effect, refer to the source page within the document.

1. Short term effects of PM_{2.5}

Regarding cardiovascular effects of short term exposure to PM_{2.5}

"Epidemiologic studies that examined the effect of PM_{2.5} on cardiovascular emergency department (ED) visits and hospital admissions reported consistent positive associations (predominantly for ischemic heart disease [IHD] and congestive heart failure [CHF]), with the majority of studies reporting increases ranging from 0.5 to 3.4% per 10 µg/m³ increase in PM_{2.5}. These effects were observed in study locations with mean 24-h average PM_{2.5} concentrations ranging from 7-18 µg/m³ (Section 6.2.10)". (p. 2-9)

“The multicity studies evaluated reported consistent increases in cardiovascular mortality ranging from 0.47 to 0.85% in study locations with mean 24-h average PM_{2.5} concentrations above 12.8 µg/m³ (Table 6-15)”. (p. 2-9)

Regarding respiratory effects of short term exposure to PM_{2.5}

“Most studies reported effects in the range of ~1% to 4% increase in respiratory hospital admissions and ED visits and were observed in study locations with mean 24-h average PM_{2.5} concentrations ranging from 6.1-22 µg/m³.” (p. 2-10)

“The multicity studies evaluated reported consistent, precise increases in respiratory mortality ranging from 1.67 to 2.20% in study locations with mean 24-h average PM_{2.5} concentrations above 12.8 µg/m³ (Table 6-15)”. (p. 2-10)

Regarding all cause mortality effects of short term exposure to PM_{2.5}

“The evaluation of multicity studies found that consistent and precise risk estimates for all-cause (nonaccidental) mortality that ranged from 0.29 to 1.21% per 10 µg/m³ December 2009 2-10 increase in PM_{2.5} at lags of 1 and 0-1 days. In these study locations, mean 24-h average PM_{2.5} concentrations were 12.8 µg/m³ and above (Table 6-15)” (p. 2-11)

2. Long term effects of PM_{2.5}

Regarding cardiovascular effects of long term exposure to PM_{2.5}

“Evidence from toxicological studies provides biological plausibility and coherence with studies of short-term exposure and cardiovascular morbidity and mortality, as well as with studies that examined long-term exposure to PM_{2.5} and cardiovascular mortality. Taken together, the evidence from epidemiologic and toxicological studies is sufficient to conclude that a causal relationship exists between long-term exposures to PM_{2.5} and cardiovascular effects.” (p. 2-12)

Regarding respiratory effects of long term exposure to PM_{2.5}

“Recent epidemiologic studies conducted in the U.S. and abroad provide evidence of associations between long-term exposure to PM_{2.5} and decrements in lung function growth, increased respiratory symptoms, and asthma development in study locations with mean PM_{2.5} concentrations ranging from 13.8 to 30 µg/m³ during the study periods (Section 7.3.1.1 and Section 7.3.2.1)... Collectively, the evidence from epidemiologic and toxicological studies is sufficient to conclude that a causal relationship is likely to exist between long-term exposures to PM and respiratory effects.” (p. 2-12)

Regarding mortality effects of long term exposure to PM_{2.5}

“The evidence for cardiovascular and respiratory morbidity due to short- and long-term exposure to PM_{2.5} provides biological plausibility for cardiovascular- and respiratory-related mortality. Collectively, the evidence is sufficient to conclude that a causal relationship exists between long-term exposures to PM_{2.5} and mortality.” (p. 2-12).

Regarding reproductive and developmental effects of long term exposure to PM_{2.5}

“The epidemiologic literature does not consistently report associations between long-term exposure to PM and preterm birth, growth restriction, birth defects or decreased sperm quality. Toxicological evidence supports an association between PM_{2.5} and PM₁₀ exposure and adverse reproductive and developmental outcomes, but provide little mechanistic information or biological plausibility for an association between long-term PM exposure and adverse birth outcomes (e.g., low birth weight or infant mortality). New evidence from animal toxicological studies on heritable mutations is of great interest, and warrants further investigation. Overall, the epidemiologic and toxicological evidence is suggestive of a causal relationship between long-term exposures to PM_{2.5} and reproductive and developmental outcomes.” (p. 2-13)

Regarding cancer, mutagenicity, and genotoxicity effects of long term exposure to PM_{2.5}

“Collectively, the evidence from epidemiologic studies, primarily those of lung cancer mortality, along with the toxicological studies that show some evidence of the mutagenic and genotoxic effects of PM is suggestive of a causal relationship between long-term exposures to PM_{2.5} and cancer.” (p. 2-13)

3. Short term effects of PM_{10-2.5}

Regarding cardiovascular effects of short term exposure to PM_{10-2.5}

“Generally positive associations were reported between short-term exposure to PM_{10-2.5} and hospital admissions or ED visits for cardiovascular causes.... The PM_{10-2.5} associations with cardiovascular hospital admissions and ED visits were observed in study locations with mean 24-h average PM_{10-2.5} concentrations ranging from 7.4 to 13 µg/m³. These results are supported by the associations observed between PM_{10-2.5} and cardiovascular mortality in areas with 24-h average PM_{10-2.5} concentrations ranging from 6.1-16.4 µg/m³ (Section 6.2.11).” (p. 2-17)

Regarding respiratory effects of short term exposure to PM_{10-2.5}

“A number of recent epidemiologic studies conducted in Canada and France found consistent, positive associations between respiratory ED visits and hospital admissions and short-term exposure to PM_{10-2.5} in studies with mean 24-h average concentrations ranging from 5.6-16.2 µg/m³ (Section 6.3.8). In these studies, the strongest relationships were observed among children, with less consistent evidence for adults and older adults (i.e., ≥ 65 years). In a large multicity study of older adults, PM_{10-2.5} was positively associated with respiratory hospital admissions in both single and copollutant models with PM_{2.5}. In addition, a U.S.-based multicity study found evidence for an increase in respiratory mortality upon short-term exposure to PM_{10-2.5}, but these associations have not been consistently December 2009 2-18 observed in single-city studies (Section 6.3.9).” (p. 2-18)

Regarding mortality effects of short term exposure to PM_{10-2.5}

“The majority of studies evaluated in this review provide some evidence for mortality associations with PM_{10-2.5} in areas with mean 24-h average concentrations ranging

from 6.1-16.4 $\mu\text{g}/\text{m}^3$. However, uncertainty surrounds the $\text{PM}_{10-2.5}$ associations reported in the studies evaluated due to the different methods used to estimate $\text{PM}_{10-2.5}$ concentrations across studies (e.g., direct measurement of $\text{PM}_{10-2.5}$ using dichotomous samplers, calculating the difference between PM_{10} and $\text{PM}_{2.5}$ concentrations). In addition, only a limited number of $\text{PM}_{10-2.5}$ studies have investigated potential confounding by gaseous copollutants or the influence of model specification on $\text{PM}_{10-2.5}$ risk estimates.” (p. 2-19)

With regards to the shape of the concentration-response curve, most of the studies evaluated in the PM ISA supported the use of no threshold log-linear models.

The World Health Organization (WHO) has developed Air Quality Guidelines (AQG) for ambient PM_{10} and $\text{PM}_{2.5}$ ³⁷. These guidelines are primarily based on published analyses of the data from selected epidemiological studies of $\text{PM}_{2.5}$ exposure, such as the Harvard Six-Cities study and the American Cancer Society study³⁸⁻⁴⁰. There are few large epidemiology studies of the effects of $\text{PM}_{10-2.5}$, yet it may also be hazardous and is the more widely-used measure of ambient air pollution: therefore, the WHO PM_{10} issued guidelines for PM_{10} that are based on those for $\text{PM}_{2.5}$, using a multiplier of 2. (In other words, the ratio of $\text{PM}_{2.5}$ to PM_{10} is assumed to be 0.5).

The WHO annual air quality guideline for $\text{PM}_{2.5}$ is 10 $\mu\text{g}/\text{m}^3$, a concentration which deciphers the lower range at which significant effects were observed in epidemiologic studies. Interim targets were also issued for countries that could not immediately meet the AQG, however these estimates are associated with higher long term mortality. Based on their mortality estimates associated with different $\text{PM}_{2.5}$ levels, the WHO estimate a 6% increase in mortality per long term 10 $\mu\text{g}/\text{m}^3$ increase in $\text{PM}_{2.5}$ above 10 $\mu\text{g}/\text{m}^3$ (and likewise 20 $\mu\text{g}/\text{m}^3$ increase in PM_{10}).

The WHO also issued short term (24 hours) air quality guidelines for $\text{PM}_{2.5}$ of 25 $\mu\text{g}/\text{m}^3$, which were based on meta-analyses of data from the US and Europe, and found consistent with data from Asia. These were based on the assumption of a 0.5% increase in mortality per 10 $\mu\text{g}/\text{m}^3$ increase in daily PM_{10} concentration (and thus 5 $\mu\text{g}/\text{m}^3$ increase in daily $\text{PM}_{2.5}$ concentration)³⁷.

Brook, et al.⁴¹ (2010) published a second scientific statement of the American Heart Association regarding the association between particulate matter and cardiovascular disease. This statement was assembled by a multi-disciplinary group of particulate matter experts that reviewed the previous AHA statement and the evidence that had been published since that statement. The group reviewed and commented on evidence regarding mortality, morbidity, association with specific diseases, biological mechanisms, and susceptible groups. A summary of the statements regarding mortality and morbidity due to PM are presented here in Brook et al.⁴¹.

With regards to time series analysis of short term exposure to PM, “The overall evidence from time-series analyses conducted worldwide since publication of the first AHA statement confirms the existence of a small, yet consistent association between increased mortality and short-term elevations in PM_{10} and $\text{PM}_{2.5}$ approximately equal to a 0.4% to 1.0% increase in daily mortality (and cardiovascular death specifically) due to a 10 $\mu\text{g}/\text{m}^3$

elevation in PM_{2.5} during the preceding 1 to 5 days (Table 2).” (p. 2338)

With regards to cohort studies on long term exposure to PM, “The overall evidence from the cohort studies demonstrates on average an approximate 10% increase in all-cause mortality per 10 µg/m³ elevation in long-term average PM_{2.5} exposure. The mortality risk specifically related to CVD appears to be elevated to a similar (or perhaps even greater) extent, ranging from 3% to 76% (Table 3). This broader estimated range in risk compared with the short-term effects observed in time series is due to several recent cohort studies that demonstrated larger cardiovascular mortality risks (e.g., >30%) than in earlier cohort observations. This may reflect superior aspects of these studies that allowed for a better characterization of the cardiovascular risk of long-term exposure, the fact that these cohorts consisted of only women, or other unclear reasons.” (p. 2341)

With regards to exposure to PM and hospitalization: “Excess cardiovascular mortality and increased rates of hospitalizations are similarly associated with day-to-day changes in PM air pollution (Tables 2 and 4). However, significant differences between geographic regions in the risk relationships have been observed, and more investigation is required to explain this heterogeneity.” (p. 2343)

Cooke et al.⁴² (2007) performed an expert elicitation with 6 European experts in epidemiology and exposure assessment to determine their views on the impact of PM_{2.5} on mortality. The experts were provided with background material, reports, and publications, and asked to estimate the effect of certain changes in PM_{2.5} concentration on mortality rates in various regions of the world. Overall, the results suggested that regulatory risk assessments underestimate the effect of PM_{2.5} on mortality. However, the effect estimates among the experts were quite variable.

Roman et al.⁴³ (2008) performed a similar expert elicitation on the impact of PM_{2.5} on mortality, however with 12 leading experts from the US. The experts generally estimated effects that were above those of the American Cancer Society study median of 0.6% per µg/m³ and below the original Six Cities median of 1.2% per µg/m³. As with the European experts, the experts generally estimated effects of PM_{2.5} and uncertainty in these estimates were generally higher than those in the EPA risk assessment for PM.

Particle number concentration

As particle size decreases, the mass of the particles becomes increasingly negligible. Ultrafine particles are a dominant source of particles in terms of PNC, yet are negligible in terms of mass. Although thought to be potent in terms of human health effects, the presence of these particles is not well-accounted for in the mass-based assessments of PM_{2.5} or PM₁₀. Recently, more studies have started assessing PNC in addition to mass in air pollution epidemiology studies.

Hoek, et al.⁴⁴ (2010) conducted an expert elicitation among toxicologists, epidemiologists, and clinicians that have well-established expertise in ultrafine particles. The purpose was to estimate the particle-response relationship and corresponding uncertainty for urban air pollution. The limited amount of information, particularly the lack of long-term studies and few studies on hospital admissions, contributed to uncertainty among the experts. Overall, the experts estimated a median (95% confidence interval (CI)) of 0.30% (0.10, 0.90%)

reduction in deaths associated with a 1,000 /cm³ decrease in UFP. For hospital admissions, the median estimates were 0.20% and 0.16% for cardiovascular and respiratory diseases, respectively (95% CI not reported).

We identified and reviewed 11 studies that specifically considered the human health effects of PNC on health. About half of these studies considered UFP specifically, whereas others considered PNC, which was typically not further defined. Based on the instrumentation used to make PNC measurements, the size ranges included in this metric were likely on the order of 20 to 1,000 nm. Overall, these studies support the findings of the expert group gathered by Hoek, et al. and suggest an increased risk of adverse cardiovascular and respiratory outcomes for increasing PNC (UFP and PNC). A summary of the results for CVD and respiratory outcomes are shown in Figures 9 and 10, respectively. Based upon the studies reviewed here, the expert elicitation estimate of 0.3% reduction in deaths per 1,000 particles/cm³ seems plausible.

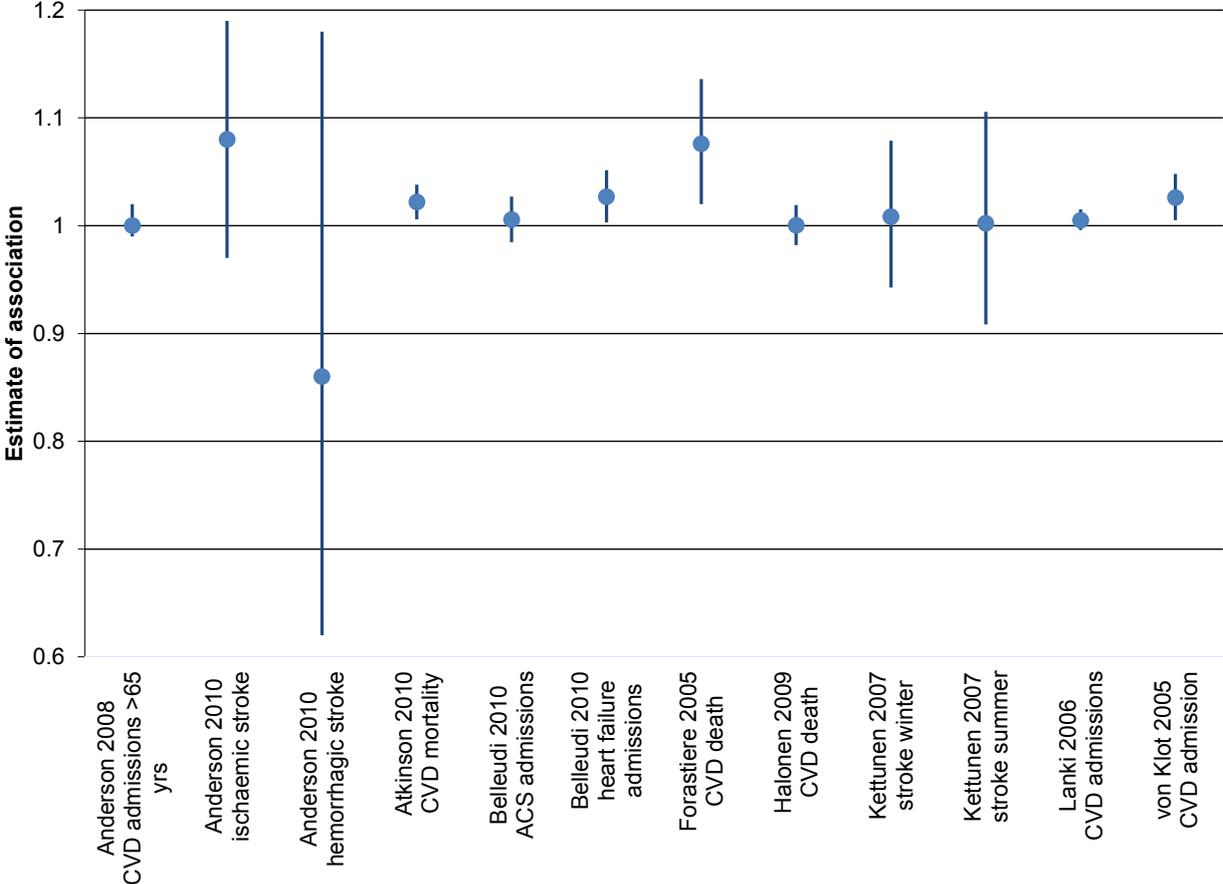


Figure 9. Estimates of association with 95% confidence interval for PNC on cardiovascular outcomes.

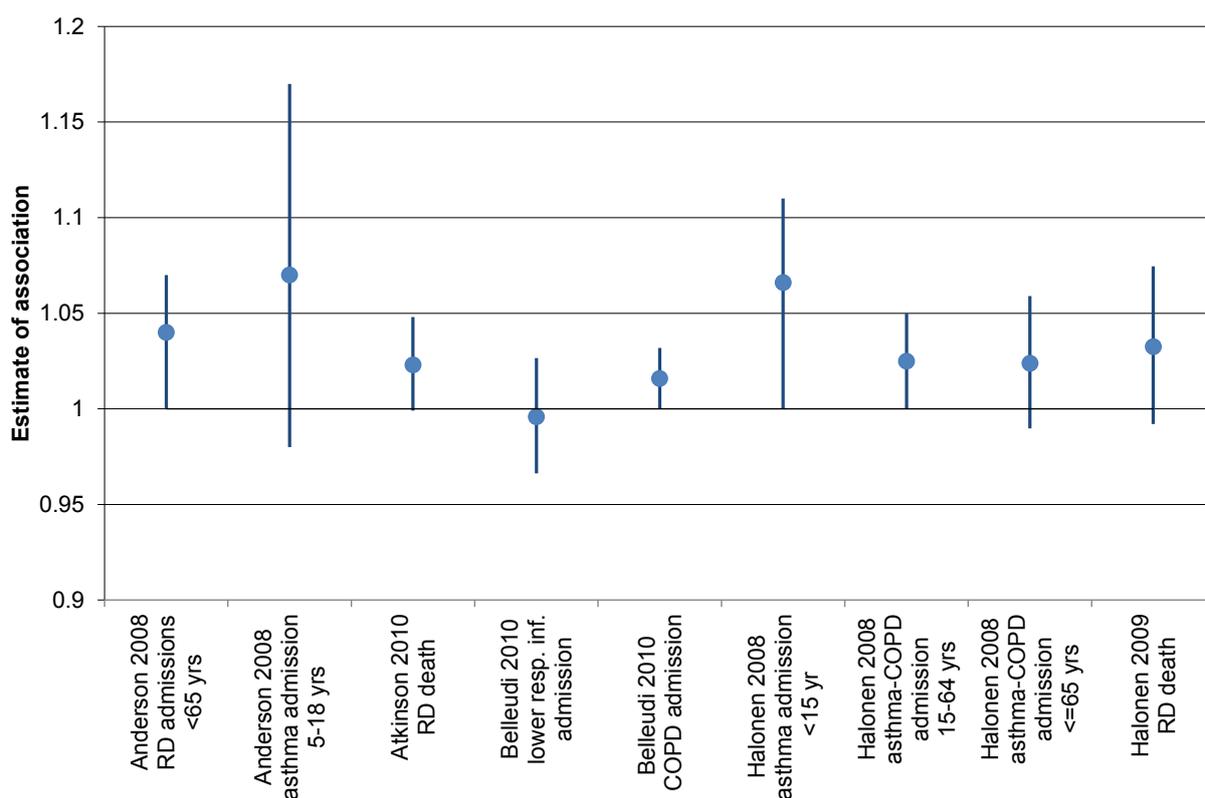


Figure 10. Estimates of association with 95% confidence interval for PNC on respiratory outcomes.

Andersen et al.⁴⁵ (2008) measured health effect in terms of respiratory or CVD hospital admissions by size and number concentration. The total number concentration (NC) was not statistically significant. Authors used interquartile ranges (IQR) for modelling ($3907/\text{cm}^3$).

Andersen et al.⁴⁶ (2010) considered effects of UFP and PM_{10} on incidence of stroke. Several types of stroke were considered, and the association was strongest (and most significant) for mild ischaemic stroke without atrial fibrillation (odds ratio, OR = 1.21). OR do not show high associations when considering all ischaemic or haemorrhagic strokes in general. Effects measured per IQR change in UFP ($3918 /\text{cm}^3$).

Atkinson et al.⁴⁷ (2010) associated PNC with daily mortality and admissions, particularly for cardiovascular diseases lagged 1-day; increases in PNC ($10,166 \text{ n}/\text{cm}^3$) were associated with 2.2% (95% CI 0.6% to 3.8%) and 0.6% (0.4% to 1.7%) increases in cardiovascular deaths and admissions, respectively. Secondary pollutants, especially non-primary $\text{PM}_{2.5}$, nitrate and sulphate, were more important for respiratory outcomes.

Belleudi et al.⁴⁸ (2010) considered the relationship between different PM fractions (PM_{10} , $\text{PM}_{2.5}$ and PNC) and admission to the hospital for various reasons. PNC showed an association only with admissions for heart failure (lag 0–5; 2.4% 0.2% to 4.7%) and COPD (lag 0; 1.6% 0.0% to 3.2%) for a $9,392 /\text{cm}^3$ increase in PNC. The effects were generally stronger in the elderly and during winter.

Forastiere et al.⁴⁹ (2005) showed that air pollution on the day of death had the strongest effect on cardiovascular death (e.g., 7.6% increase [95% CI, 2.0–13.6%]) for an interquartile range of PNC, 27,790 particles/cm³.

Halonen et al.⁵⁰ (2008) assessed relationship between respiratory outcomes and particulate matter by size range, including nucleation (<0.03 µm), Aitken (0.03–0.1 µm) and accumulation (0.1–0.29 µm) mode particles. For the purpose of this study, the Aitken mode was considered as UFP (results showed higher significance than for nucleation mode). Observations were per IQR increase in particles (5760 /cm³).

Halonen et al.⁵¹ (2009) considered association between hospital admissions for CVD and RD and particle number. They found that there was a suggestion of an association of hospital admissions for arrhythmia with Aitken mode particles and PM_{2.5} from traffic. Otherwise few associations were observed between various sizes and types of particles for either cardiovascular admissions or mortality. In contrast, most particle fractions had positive associations with admissions for pneumonia and asthma-chronic obstructive pulmonary disease (COPD). For the purpose of this review, the Aitken mode was considered as UFP (results showed higher significance than for nucleation mode). Observations were per IQR increase in Aitken mode particles (2467 /cm³).

Ketunnen et al.⁵² (2007) evaluated the association between stroke and UFP. They found significant associations in the warm season, but not in the cold season. Percent changes in stroke were assessed per 7330 /cm³ increase in particle concentration.

Lanki et al.⁵³ (2006) found an association of the same day PNC levels with acute myocardial infarction (AMI). However, associations were only observed in the three cities with hospital discharge registers (v. general AMI registries) where power for city-specific analyses was higher. Effects of air pollution were more pronounced during the warm than the cold season.

Stolzel et al.⁵⁴ (2007) found statistically significant associations between elevated ultrafine particle (UFP; diameter: 0.01–0.1 µm) number concentration and total as well as cardio-respiratory mortality, each with a 4 days lag. The relative mortality risk (RR) for a 9748 cm³ increase in UFP NC was RR: 1.029 and its 95% CI 1.003–1.055 for total mortality. For cardio-respiratory mortality they found RR: 1.031, 95% CI: 1.003–1.060. This study is not included in Figures 9 and 10 because it did not consider CVD and RD separately.

Von Klot et al.⁵⁵ (2005) evaluated cardiac readmissions in association with PNC. They found that cardiac readmissions increased in association with same-day estimated PNC per 10000 particles/cm³. Pooled effect estimates for angina pectoris and myocardial infarction readmissions were comparable

Chemical composition

The analysis of the chemical composition in air monitoring data is relatively recent and there is not a large body of data accumulated yet for detailed analysis.

Volcanic ash

Volcanic ash can have high crystalline silica content, which is associated with silicosis, a scarring of lung tissue. Additionally, the particles can have high surface Fe²⁺ content,

which is associated with the formation of free radicals. Periods of high exposure to volcanic ash are typically not chronic.

Acute respiratory symptoms (e.g., irritation) but no long term effects have been observed in the few (and rather small) studies conducted to date⁵⁶.

Wildfire smoke

As with volcanic ash, most exposures to high levels of wildfire particulate are of an acute nature. Studies have shown significant increases in acute respiratory symptoms (e.g., asthma, rhinitis) and smaller but significant increases for more chronic respiratory diseases in vulnerable populations (i.e., bronchitis and COPD in certain age groups)⁵⁷⁻⁵⁹.

Source apportionment studies

Source apportionment studies use defined emissions profiles from various sources to identify the portion of ambient PM that can be attributed to these sources. The emission profiles can be defined a priori or using statistical analysis of the emission profiles. These studies link health effects to a source, not to a particular component of PM. Studies reviewed so far have considered outcomes such as cause-specific emergency room visits, non-accidental mortality, and changes in cardiac blood factors and sources such mobile source PM_{2.5}, biomass combustion PM_{2.5}, secondary sulphate PM_{2.5}, and traffic speed changes⁶⁰⁻⁶².

6.3 Chemical composition of PM

In chapter 5.1 it was reported that the analysis of the chemical composition in air monitoring data is relatively recent and there is not a large body of data accumulated yet for detailed analysis. On account of this, the literature search on chemical composition was intensified and a number of studies was found that allows even to perform statistical analysis in some cases. However, for most substances statistical analysis is inappropriate due to a lack of data. The search resulted in a total of 16^{60, 62-76} studies which provide RRs to almost 40 different substances.

6.4 Evaluation of the Literature for PNC of UFP and precursor gases of secondary particles (SO_x, NO_x and CO)

The literature review resulted in 28 studies^{45-55, 63, 65, 66, 77-89} providing information for PM₁₀, PM_{2.5}, PNC and (precursor gases of) secondary particles SO_x, NO_x and CO. Table A1 in the Annex provides single data points of the RR and the CI for PM₁₀, PM_{2.5}, PNC, SO_x, NO_x and CO. Table 4 summarizes the total number of data points read out from the 28 studies for all different health endpoints. The blue fields indicate the total number of data points for a single substance (e.g. PNC vs. PNC=40 implies that 40 data points were found for PNC). Numbers in the right upper part of the blue fields indicate the number of data pairs for two substances (e.g. PM_{2.5} vs. PNC=29 implies that there are 29 paired data points for PNC and PM₁₀ and paired means that both elements of the pair stem from the same study). In the left lower part the results for sample size from the power analysis are provided. Analysis of the mean values from PNC and PM_{2.5} with paired sample t-test contains 29 data points. Based on the mean values and the standard deviation for PNC

and PM_{2.5} power analysis calculates a minimal number of data points required of 208 in order to detect likely an effect (based on Power P=0.8 and a significance level α of 0.05.).

	PM ₁₀	PM _{2.5}	PNC	SO _x	NO _x	CO
PM ₁₀	68	50	40	27	50	36
PM _{2.5}	141	55	29	23	37	23
PNC	36	208	40	8	26	19
SO _x	179	38	30	32	29	16
NO _x	371	307	120	1107	55	35
CO	2343	15	75	453	723469	36

Table 4. Data points found and number of data points required using power analysis. Blue fields indicate the total number of data points for a single substance. Number of paired data points for the 6 substances are shown in the upper right part and sample size required in order to get statistically significant effects using power analysis are shown in the lower left part.

Figure 11 depicts a comparison between all PM size classes and the (precursor gases of) secondary PM for total effects on human health (CVD, RD and lung cancer). The mean values are calculated from paired values so that differences in testing conditions are eliminated.

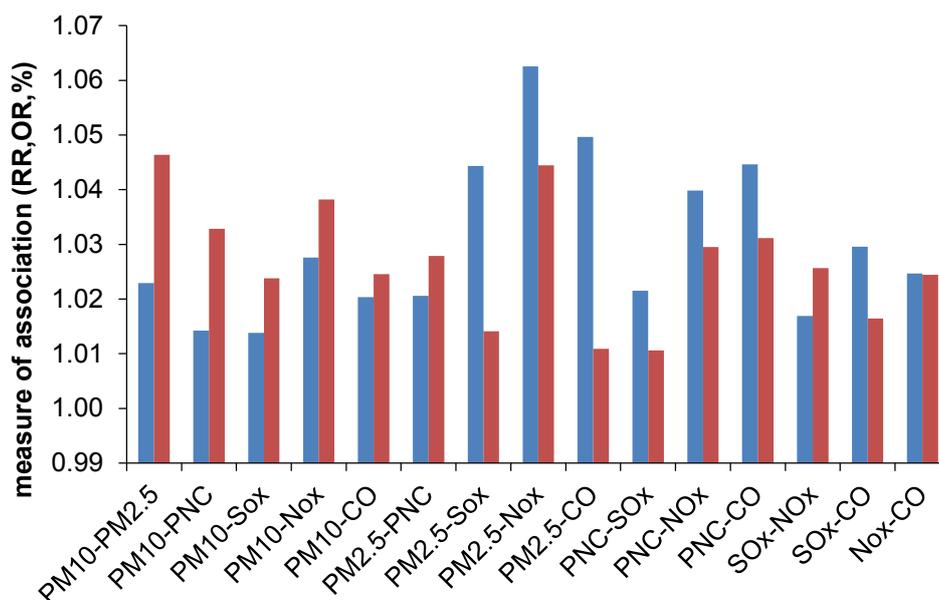


Figure 11. Mean of PM size classes and the secondary PM.

PM₁₀ causes less human health effects than any other substances in the comparisons above. In contrary, comparing PNC with all other substances shows that PNC has the highest damage potential. PM_{2.5} has a smaller damage potential than PNC but a higher potential than any other substance.

Table 5 provides an overview of the results from statistics using pairwise t-test. The numbers in the left lower part of the table refer to P-values (level of significance P=0.05). Statistically significant differences (red) and results that tend to be different (green) are highlighted.

	PM ₁₀	PM _{2.5}	PNC	SO _x	NO _x	CO
PM ₁₀	68	50	40	27	50	36
PM _{2.5}	0.003	55	29	23	37	23
PNC	0	0.18	40	8	26	19
SO _x	0.058	0.024	0.179	32	29	16
NO _x	0.081	0.208	0.052	0.653	55	35
CO	0.679	0.065	0.008	0.614	0.982	36

Table 5 Data points found and statistical analysis. Number of data points for the 6 substances (upper right part) and P-values from paired sample t-test (lower left part) without α error correction. Red highlighted numbers refer to statistically significant comparisons ($P < 0.05$) and green highlighted values indicate a tendency to a significance ($P < 0.1$).

Comparing PNC and PM₁₀, the effects from PNC (3.3%) is more than twice as high than the effect of PM₁₀ (1.4%) on a level of significance $P < 0.001$. Further, the mean risk of PM_{2.5} (4.6%) is double the risk of PM₁₀ (2.3%; $P = 0.003$). The number of data points available is considerably smaller ($n = 50$) than the number of data points required as determined using power analysis ($n = 141$). In addition, both examples show a level of significance much higher than what could be expected (expected could be $P = 0.05$ based on power analysis) although the number of studies, at least in the case of PM_{2.5}, is much smaller than what power analysis computes. Both, the level of significance and the number of studies available are strong indications of having found an essential effect of particle size. The same discussion could be accomplished for several other examples too (e.g. PNC vs. CO, etc.).

The results seem to be strong. However, there are many differences between the studies (Table 6) which might affect the results. Such differences are for example the study population, exposure concentration, the time frame of the study, etc.

Author	Year	PM ₁₀			PM _{2.5}			PNC			SO _x			NO _x			CO		
		Mean µg/m ³	IQR µg/m ³	n #	Mean µg/m ³	IQR µg/m ³	n #	Mean #/cm ³	IQR #/cm ³	n #	Mean	IQR	n #	Mean	IQR	n #	Mean	IQR	n #
Anderson	2008	24	13	1169	10	5	453	8116	3907	578				15	9	1245	290	120	1230
Anderson	2010	27.1	13.5	1394				6365	3918	939				15.3	8.8	1418	270	120	1375
Atkinson	2010	22	14	1921	15	11	1942	18769	10456	1725									
Beelen	2008																		
Belleudi	2010	39.3	18	1704	22.8	12.8	1448	37456	26180	1433									
Breitner	2009	38	28.3	3802	25.6	30.4	2516	12910	8685	2359				35.2	18.5	3467	0.63	0.54	3761
Dadvand	2011																		
Forrestriere	2005	52.1	29.7					51669	27790		5.2	4.4		71.4	18.5		2.4	1.2	
Gehring	2006	43.7	7.1											39	16				
Halonen	2008	9.9	7.2	2392	9.5	6.2	2424	8203	5760	2436				28.2	14.2	2550	0.5	0.3	2550
Halonen	2009	9.9	7.2	2392	9.5	6.2	2424	8203	5760	2436				28.2	14.2	2550	0.5	0.3	2550
Jalaludin	2006	16.8	7.8		9.5	8.9					1.07	0.75		23.2	9.3		820	690	
Kettunen	2007	16.3	13.8	1454	8.2	6.7	1419	8986	7330	1452				27.6	14.6	1482	0.5	0.2	1482
Lanki	2006																		
Leitte	2011	120	90	1036				22000	11000	818	87	100	1036	63	40	1036			
Lipfert	2006	16	5.1		14.3	3.2													
Naess	2006																		
Peel	2005	27.9	12.3	2703	19.2	8.9		38000	40700										
Peel	2007																		
Pope	2002	28.5	5.9		17.7	3.7					6.5	2.8		21.4	7.1		1100	400	
Rosenlund	2006																		
Samoli	2011	43.9	21.6								16.8	13		84.8	37.3				
Sarnat	2008																		
Slaughter	2005																		
Stölzl	2007	32.3	23.8	1976	21.7	16.3	1994	13491	9748	2000				32.4	17.2	2154	0.47	0.34	2187
Tolbert	2007	26.6	16.3	4264							14.9	16	4358	43.2	23	4351	1600	1200	4275
Villeneuve	2006	24.2	16	1480	8.5	6.3	1440				2.6	3	3616	24	13.5	3650	800	500	3650
von Klot	2005																		
Mean		32.6	18.5	2130	14.7	9.7	1784	19514	13436	1618	not calculated because of different units used in literature: ppb, ppm, µg/m ³ , mg/m ³								
SD		24.2	18.8	983	6.1	7.2	667	14935	11675	690									
N		19	19	13	13	13	9	12	12	10									

Table 6. Exposure variability between studies. Differences between studies with respect to exposure concentration, number of days the pollutant was measured and the interquartile increase of the pollutant associated with the human health effect.

In order to take account for uncertainties arising from different setup in studies, all used studies could be weighted according to the relevant parameters, e.g. sample size, time frame, etc.

6.5 Inverse Variance method: Weighting studies for meta-analysis purpose

A simple method – inverse variance method – is used to calculate the weights required for further analysis. Optimal weights for meta-analysis are:

$$w = \frac{1}{SE^2}$$

Where SE (standard error) can be calculated with the following formula

$$CI_{95\%} = \bar{x} \pm SE = \bar{x} \pm \frac{\sigma}{\sqrt{N}}$$

The mean effect size (ES) from all studies can be calculated as

$$\overline{ES} = \frac{w \times ES}{w}$$

For odds ratio (OR) and relative risks (RR), ES is usually logarithmically transformed, meaning one should use:

$$\log \overline{ES} = \frac{w \times \log ES}{w}$$

Statistical analysis of the weighted study results indeed lead to no longer significant differences between PM₁₀, PM_{2.5} and UFP. However, PM_{2.5} tends to be significantly different from PM₁₀ (P=0.07). But the results have to be interpreted carefully. For the weighting of the results the 95% CI is the starting point. The CI depends on one hand on the study population, but other influencing parameters are also used for the calculation of the CI (personal communication with Prof Nino Künzli, Swiss TPH, Basel), in particular the number days on which PM₁₀, PM_{2.5} and UFP are measured.

The 95% CI (of each data point provided in the literature studies) provides insight into the explained variance of the compared substances. Table 7 shows the mean values, CI and the number of data points (N, number of days on which the emissions was measured) available for the calculation of the CI for PM₁₀ and PNC. Only 24 CI were provided of the total number of 40 paired samples of PM₁₀ and PNC.

	PM ₁₀			PNC				
	Mean	CI	N	Mean	CI	N	d.CI	d. N
Anderson	1.03	0.02	1169	1.03	0.02	578	0	591
	1.06	0.035	1169	1.04	0.035	578	0	591
	1.02	0.095	1169	1.07	0.095	578	0	591
Anderson	1.02	0.055	1394	1.08	0.11	939	-0.055	455
	1.01	0.065	1394	1.08	0.125	939	-0.06	455
Atkinson	1.001	0.008	1921	1.022	0.016	1725	-0.008	196
	1.003	0.005	1921	1.009	0.012	1725	-0.007	196
	1.02	0.012	1921	1.012	0.023	1725	-0.011	196
	1.008	0.01	1921	0.985	0.015	1725	-0.005	196
Belleudi	0.9945	0.01705	1704	1.0056	0.0211	1433	-0.00405	271
	1.0319	0.02725	1704	1.0268	0.0242	1433	0.00305	271
	1.021	0.03345	1704	0.9959	0.0302	1433	0.00325	271
	0.9819	0.02755	1704	0.9985	0.0256	1433	0.00195	271
Breitner	1.006	0.0255	3802	1.055	0.0445	2359	-0.019	1443
	1.017	0.027	3802	1.063	0.0475	2359	-0.0205	1443
Halonen	1.0255	0.02825	2392	1.0606	0.02975	2436	-0.0015	-44
	1.027	0.02525	2392	1.0249	0.0246	2436	0.00065	-44
Kettunen	0.9944	0.02805	1454	1.0083	0.06815	1452	-0.0401	2
	1.1089	0.1043	1454	1.0848	0.1011	1452	0.0032	2
Leitte	1.003	0.03	1036	1.03	0.055	818	-0.025	218
	1.003	0.04	1036	1.07	0.085	818	-0.045	218
	1.02	0.035	1036	1.06	0.05	818	-0.015	218
Stölzl	1.004	0.0245	2190	1.042	0.028	2000	-0.0035	190
	1.007	0.0265	2190	1.031	0.0285	2000	-0.002	190

Table 7. Mean, confidence intervals (CI), number of days (N) for PM₁₀ and PNC and the difference between the CI and N for PM₁₀ and PNC.

The calculation of the CI includes the number of days on which the emission was measured. Unfortunately, this number varies considerably between PM₁₀ and PNC. A high amount of data points available lead to small CI, few data points available lead to higher CI (personal communication with Prof Nino Künzli, Swiss TPH, Basel).

There is one case (red highlighted in Table 7) in which:

a) CI of PNC > CI of PM₁₀, although b) N of PNC > N of PM₁₀. This example does not support the hypothesis that the CI for each size class is smaller than CI for PM₁₀.

However, there are 7 cases (green highlighted in Table 7) in which:

a) CI of PNC < or = CI of PM₁₀, although b) N of PNC < or = N of PM₁₀. These examples support the hypothesis that the CI for split size classes is smaller than CI for PM₁₀.

This fact and the strong results from the power analysis together with the statistical results for PM₁₀, PM_{2.5} and PNC lead to the decision to use the effects of size found in the literature study, although I failed to prove significant differences between the 3 size classes after weighting of the studies.

6.6 Evaluation of the Literature for the chemical composition of PM

Table 8 provides an overview over the data points gathered from 16 studies of all substances found in the literature and the number of all possible pairs of data (one pair of data is generated when values for 2 substances are provided in the same study). The number of data points is rather small for most substances. Comprehensive statistical analysis of such a dataset provides few statistically significant results, since the differences between the studies are too substantial, e.g. variety of exposure concentration between studies, timeframe, etc. (see Table 7 for the variation of exposure to PM₁₀, PM_{2.5} and PNC between studies). Based on the findings from the weighting of studies to find differences between size classes it is not reasonable to further analyse the data available for chemical substances using the inverse variance method.

	Black smoke	PM 2.5	EC, BC	OC	Zinc	Potassium	Silicium	Selenium	HCO	Vanadium	Nickel	Nitrate	Chlorine	Sodium	Copper	Lead	Ammonia	Aluminium	Iron	Arsenic	Calcium	Sulfate	Magnesium	Titanium	Sulfur	Manganese	Chromium	Bromine	Barium	Phosphorus	Biomass/wood comb.	oil/coal comb	crustal	sea salt	vehicle	road dust	Sulfur (S+P from regional sources)	CU smelter				
Black smoke	3	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0		
PM2.5		11	7	7	9	6	5	1	0	0	1	1	7	0	0	7	1	0	1	3	3	0	7	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
EC, BC		0.2	22	18	17	16	15	9	3	2	7	8	11	7	10	14	9	4	9	11	3	7	11	1	5	6	4	4	3	4	4	4	4	4	0	4	4	4	4	4	0	
OC		0.1	0.22	18	13	12	11	5	3	2	3	4	11	3	6	10	5	4	5	7	3	3	11	1	1	2	4	4	3	0	0	0	0	0	0	0	0	0	0	0	0	
Zinc		0.4	0	0.9	19	16	15	9	0	0	7	8	9	7	8	14	9	2	9	11	5	7	9	1	5	6	4	4	3	4	4	6	4	2	4	6	4	4	4	0		
Potassium			0.03	0.05	0.29	16	12	8	0	0	6	7	8	7	8	13	8	2	8	10	2	7	8	1	5	6	3	3	3	4	4	4	4	4	0	4	4	4	4	0		
Silicon			0.06	0	0.95	0.04	17	9	0	0	7	8	7	7	10	10	9	4	9	11	5	7	7	1	5	6	4	4	3	4	4	6	4	2	4	6	4	4	0			
Selenium			0.01		0.06	0.31	0.03	9	0	0	5	7	1	6	6	7	7	0	7	7	1	6	1	0	4	6	3	2	4	4	4	4	4	4	0	4	4	4	4	0		
HCO									3	2	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	
water soluble metals										2	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	
Vanadium			0.14		0.48		0.66			8	7	3	5	6	6	7	2	7	7	3	5	3	1	5	4	2	2	1	4	4	4	4	4	0	4	4	4	4	4	0		
Nickel			0.01		0.63	0.45	0.86	0.218		0.42	10	2	7	7	8	9	1	8	8	2	7	2	1	5	7	3	3	2	4	4	4	4	5	1	4	5	4	4	0			
Nitrate		0.1	0.1	0.4	0.4	0.28	0.26				11	1	4	8	3	4	3	5	3	1	11	1	1	0	2	2	1	0	0	0	0	0	0	0	0	0	0	0	0	0		
Chlorine			0		0.26	0.7	0.54				0.06		7	7	7	7	1	7	7	1	7	1	1	5	6	2	2	2	4	4	4	4	4	4	0	4	4	4	4	0		
Sodium			0.01		0.04	0.71	0.41				0.16	0.24	0.69	10	7	8	4	8	8	2	7	4	1	5	6	3	3	4	4	4	4	4	4	4	0	4	4	4	4	0		
Copper		0.2	0.01	0.64	0.79	0.16	0.92	0.009			0.84	0.26	0.24	0.1	14	8	1	8	10	2	7	8	1	5	6	3	3	2	4	4	4	4	4	0	4	4	4	4	0			
Lead			0.16		0.13	0.03	0.15	0.004		0.91	0.57		0.11	0.06	0.07	10	2	9	9	3	7	3	1	5	7	4	4	3	4	4	4	4	5	1	4	5	4	4	0			
Ammonia																	4	2	2	2	1	4	1	1	0	1	1	1	0	0	0	0	0	0	0	0	0	0	0	0		
Aluminium			0.15		0.52	0	0.03	0.015		0.78	0.9		0.29	0.18	0.58	0.37		9	9	3	7	3	1	5	6	4	4	3	4	4	4	4	4	0	4	4	4	4	0			
Iron			0.07	0.17	0.73	0.02	0.55	0.006		0.69	0.83		0.56	0.43	0.86	0.04		0.07	11	3	7	5	1	5	6	4	4	3	4	4	4	4	0	4	4	4	4	4	0			
Arsenic																			5	1	3	1	1	1	0	2	2	1	0	0	2	0	2	0	2	0	2	0	0	0		
Calcium			0.51		0.33	0.03	0.1			0.54		0.27	0.22	0.31	0.65		0.39	0.08		7	1	1	1	5	6	2	2	2	4	4	4	4	4	0	4	4	4	4	0			
Sulfate		0.1	0.24	0.2	0.32	0.83	0.71			0.3	0.33				0.37																											
Magnesium																																										
Titanium																																										
Sulfur																0.06																										
Manganese																																										
Chromium																																										
Bromine																																										
Barium																																										
Phosphorus																																										
Biomass/wood comb.																																										
oil/coal comb																																										
crustal																																										
sea salt																																										
vehicle																																										
road dust																																										
Sulfur (S+P from regional sources)																																										
CU smelter																																										

Table 8. Number of data points (blue fields), number of paired samples (upper right part) and P-values from paired sample t-test (lower left part).

The data don't include a error correction. Green: statistically significant (P<0.01), yellow: (P<0.05) and red: tendency to a significance (P<0.1).

6.7 PAH, Dioxin and other potentially toxic substances

Unfortunately, no single epidemiological study was found which provided information to PAH and dioxin/furan or polychlorinated biphenyls (PCB). With regard to the chemical composition the lack of information about PAH and dioxin is certainly the most severe gap. However, other potentially highly toxic substances, such as heavy metals (hexavalent chromium or lead), metalloids (arsenic) or non-metals (selenium) are not assessed well enough to characterise PM in an appropriate way.

The results for the chemical composition found in the literature are too weak and can't be used for a comprehensive chemical characterisation of PM. The chemical characterisation of PM has to be based on other data than on those from epidemiological studies.

Alternatively, useful data were found in a US database⁹⁰ which provides unit risk factors (URF) for lung cancer disease and reference concentrations (RFC) for cardiopulmonary disease (CPD). The comprehensive database contains URF and RFC of about 240 chemicals found in ambient air. All substances are listed in the Annex (Table A2).

6.8 Unit risk factor and reference concentration

The URF is defined as follows: The upper-bound excess lifetime cancer risk estimated to result from continuous exposure to an agent at a concentration of $1 \mu\text{g}/\text{m}^3$ in air. The interpretation of inhalation unit risk would be as follows: if unit risk = 2×10^{-6} per $\mu\text{g}/\text{m}^3$, 2 excess cancer cases (upper bound estimate) are expected to develop per 1'000'000 people if exposed daily for a lifetime to $1 \mu\text{g}$ of the chemical in $1 \mu\text{g}/\text{m}^3$ air.

The RFC is an estimate (with uncertainty spanning perhaps an order of magnitude) of a continuous inhalation exposure to the human population (including sensitive subgroups) that is likely to be without an appreciable risk of deleterious effects during a lifetime. It can be derived from a NOAEL (No Observed Adverse Effect Level), LOAEL (Lowest Observed Adverse Effect Level), or benchmark concentration, with uncertainty factors generally applied to reflect limitations of the data used. The RFC is generally used in EPA's noncancerous health assessments.

7 Description of PM in ambient air based using size classes and chemical composition

A prerequisite for the selection of chemical and physical parameters to characterise damage to human health is that the parameter is measured in ambient air. Thus, a characterisation of European ambient air had to be achieved accounting on one hand on all substances with high shares on ambient air composition, but then also highly toxic substances found in extremely low concentrations.

7.1 Main components

A study from Putaud et al.⁹¹ serves as a comprehensive data basis. This article synthesizes data on aerosol physical and chemical characteristics, which were obtained in European aerosol research activities at free-troposphere. It contains data from 60 natural, rural, near-city, urban, and kerbside sites over the past decade. The data include PM₁₀ and PM_{2.5} mass concentrations, aerosol PNC, and size segregated chemical main composition (carbon black, organic matter, nitrate, ammonia, sulphate, sea salt, mineral dust) for PM₁₀ and PM_{2.5}.

The study did not provide chemical composition of UFP. A review from Chow & Watson⁹² combined with few other studies⁹³⁻⁹⁶ provides the required literature and data on the main chemical components for UFPs.

Wikipedia describes mineral dust as to be composed mainly on the oxides (SiO₂, Al₂O₃, FeO, Fe₂O₃, CaO, and others) and carbonates (CaCO₃, MgCO₃) that assemble the Earth's crust⁹⁷. Global mineral dust emissions are estimated 100-500 millions of tons per year, of which the largest part is attributed to deserts. Although this aerosol class is usually considered of natural origin, it is estimated that about 30% of the mineral dust load in the atmosphere could be ascribed to human activities through desertification and land misuse. Despite the rather high contribution of mineral dust on the total PM mass, mineral dust is assumed to have a low toxic potential on human health. Thus, it is assumed that all substances listed above have equal shares on the total amount of mineral dust.

Similar to mineral dust, sea salts are assumed to have low toxic potential but a significant share on the total amount on ambient PM. Sea salt is produced from evaporating sea spray, which is a spray of sea water that forms when ocean waves crash. The main components of sea salt⁹⁸ are listed in Table 9.

Element	Relative concentration
Chloride (Cl ⁻)	55.03%
Sodium (Na ⁺)	30.59%
Sulfate (SO ₄ ²⁻)	7.68%
Magnesium (Mg ²⁺)	3.68%
Calcium (Ca ²⁺)	1.18%
Potassium (K ⁺)	1.11%
Bicarbonate (HCO ₃ ⁻)	0.41%
Bromide (Br ⁻)	0.19%
Borate (BO ₃ ³⁻)	0.08%
Strontium (Sr ²⁺)	0.04%
Miscellaneous constituents	0.01%
Total	100.00%

Table 9. Composition of sea salt.

7.2 Trace elements

Trace elements⁹⁹ were assigned using the results from Hüglin & Gehrig¹⁰⁰ who chemically characterised PM_{2.5}, PM₁₀ and coarse particles at urban, near-city and rural sites in Switzerland. Chemical composition from other locations rounded up the chemical characterisation of trace elements, in particular trace elements with respect to UFPs (Erfurt¹⁰¹, Netherland¹⁰², Los Angeles^{93, 95}).

7.3 Organic matter

Organic matter (OM) is an important fraction of the atmospheric aerosol that, as result of analytical difficulties and the large number of individual compounds present, is still poorly characterised. In contrast to sea salt or mineral dust OM is considered as being highly toxic. This might be the truth for some components (dioxin, furan, PCB) of OM, but probably not for all substances. Therefore OM should be split in different groups.

Regardless on functional compounds attached to the chemical skeleton, organic substances can be split in aliphatic and aromatic substances. Aromatics are further divided into monocyclic and polycyclic aromatic substances. Aliphatic substances can be further split into alkanes/alkenes/alkynes, aldehydes, ketones, alcohols and acids. The required ambient levels of each of the components above are derived from 3 European studies¹⁰³⁻¹⁰⁵, a US study¹⁰⁶ and an Asian study¹⁰⁷.

7.4 Dioxins and dioxin-like compounds

Dioxins and dioxin-like compounds (DLC) are by-products of various industrial processes, and are commonly regarded as highly toxic compounds that are environmental pollutants and persistent organic pollutants. DLCs have a common chemical skeleton build up on two benzene rings. The two rings are directly chemically connected (PCB) or connected via one (PCDF) or two (PCDD) oxygen atoms. In addition, all DLCs have at least one or several hydrogen atoms substituted by halogen elements (chlorine, bromine).

PCDD and PCDF are two groups of persistent, semi-volatile and toxicologically significant trace organic contaminants. They enter the environment in ultra-trace amounts from various combustion sources and as chemical impurities in a range of manufactured

organo-chlorine products. Lohmann & Jones¹⁰⁸ provided a comprehensive, critical review of the levels, behaviour and processes affecting dioxins and furans in ambient air. Data from 25 kerbside, urban and rural sites from all over Europe were used for the characterisation of dioxins and furans in ambient European air.

Other widely discussed persistent, organic pollutants are PCBs. Current atmospheric levels of PCBs in the environment are due to mainly anthropogenic emissions. The production and disposal or accidental releases of products or materials containing PCBs, volatilization from environmental reservoirs (e.g., sea, soil), or unwanted formation of some congeners from e.g. combustion processes are the major source of PCBs. Jaward et al.¹⁰⁹ characterised PCBs in ambient air at 71 sites in 22 countries all over Europe.

8 Modelling Fate, Exposure, Effects and Damage

The impact to human health assessed due to PM emissions depends on the emission profile, the fate of PM in the atmosphere, the exposition of the population to the emission (intake and uptake fraction), the human health effects resulting from the exposition and the damage calculated in DALY. The CF is the number resulting from the entire impact assessment model and directly links the emissions to the impact. The model for the impact assessment can be written as:

$$\text{CF (DALY)} = f(\text{Emissions; Fate; Exposure, HH-Effect, Damage})$$

8.1 Modelling the background concentration

The modelling of the emissions is content of the life cycle inventory modelling. LCA generally applies “marginal change” approach. Thus, It is assumed that an additional amount of a certain stressor (e.g. PAH) introduces very small changes on top of a ceteris paribus background situation. The change in impact per unit amount of additional “release” represents the relative importance of the stressor toward a specific area of protection, i.e., human health. The reasoning behind working with marginal changes is that in LCA the focus is on small changes via the concept of the functional unit (e.g. 1 MJ heat from wood combustion)¹¹⁰. For most impact categories, fate and exposure factors are derived with linear environmental models. Hence, background concentration of all components the impact assessment takes into account have to be determined.

From the literature review on the characterisation of particles in ambient air related to three different site specifications (rural, urban, kerbside) the following mean concentration for PM₁₀, PM_{2.5} and UFP were found (Table 10):

	PM ₁₀ µg/m ³	PM _{2.5} µg/m ³	UFP 10 ³ Particles/cm ³
Kerbside	33.8	20.4	17.9
Urban	24.8	17.7	9.6
Rural	11.8	8.9	3.6

Table 10. Mean ambient European PM concentration at three different sites.

This rough characterisation based on particle size has to be broken down into a much more detailed level in order to find the required shares of the chemically relevant species of ambient European air. Table 11 gives an overview on the most relevant chemical substances and their contributions. The whole list of all chemically identified substances for this study is provided in the Annex (Table A3).

Ambient air concentration of chemical substances									
Pollutant unit site	PM ₁₀ µg/m ³			PM _{2.5} µg/m ³			UFP Particles/cm ³		
	rural	urban	kerbside	rural	urban	kerbside	rural	urban	kerbside
Carbon Black	9.8E-01	1.7E+00	5.4E+00	8.9E-01	1.7E+00	4.1E+00	2.7E+02	7.2E+02	1.4E+03
Nitrate	1.1E+00	5.1E+00	4.2E+00	5.5E-01	3.6E+00	2.6E+00	1.9E+02	4.9E+02	9.2E+02
Ammonia	9.8E-01	2.4E+00	1.7E+00	8.9E-01	2.1E+00	1.7E+00	1.3E+02	3.6E+02	6.7E+02
Sulfates	3.1E+00	4.4E+00	4.2E+00	3.2E+00	3.6E+00	3.1E+00	1.9E+02	5.2E+02	9.6E+02
Asbestos	6.1E-08	1.3E-07	1.7E-07	6.5E-08	1.3E-07	1.5E-07	2.6E-05	7.0E-05	1.3E-04
Sea salt total	1.3E+00	1.4E+00	1.3E+00	3.3E-01	6.4E-01	2.4E-01	1.4E+02	3.7E+02	6.8E+02
Mineral dust total	9.4E-01	1.7E+00	4.5E+00	3.2E-01	8.5E-01	1.1E+00	4.1E+02	1.1E+03	2.0E+03
Trace elements total	7.0E-01	1.3E+00	3.4E+00	2.4E-01	6.4E-01	8.2E-01	3.1E+02	8.2E+02	1.5E+03
Organic Matter total	2.6E+00	6.8E+00	9.2E+00	2.5E+00	4.7E+00	6.9E+00	2.0E+03	5.2E+03	9.8E+03
Dioxins, PCDD	1.2E-05	2.1E-05	2.8E-05	1.0E-05	2.2E-05	1.9E-05	2.8E-02	7.4E-02	1.4E-01
Furane, PCDF	1.6E-05	2.8E-05	3.8E-05	1.3E-05	2.9E-05	2.6E-05	3.7E-02	9.9E-02	1.9E-01
PCB	2.0E-04	3.4E-04	4.7E-04	1.7E-04	3.6E-04	3.2E-04	4.6E-01	1.2E+00	2.3E+00
Aliphatic total	2.5E+00	6.4E+00	8.6E+00	2.4E+00	4.4E+00	6.5E+00	1.8E+03	4.9E+03	9.1E+03
Aromatics total	1.7E-01	4.4E-01	5.9E-01	1.6E-01	3.0E-01	4.5E-01	1.3E+02	3.4E+02	6.3E+02
Aromatics (monocyclic)	1.2E-01	3.2E-01	4.4E-01	1.2E-01	2.2E-01	3.3E-01	9.3E+01	2.5E+02	4.6E+02
PAH	4.5E-02	1.2E-01	1.6E-01	4.4E-02	8.0E-02	1.2E-01	3.4E+01	9.0E+01	1.7E+02

Table 11. Overview on the most relevant chemical substances and their contributions.

Although there is a very good literature base available for the ambient air PM, due to the complexity of PM we had to take few assumptions (Table 12):

Assumptions and limitations for the background concentration

- Mineral dust: all single components are assumed to contribute with the same mass share.
- OM: The further split into components (such as ketones, aldehydes, acids, etc.) is arbitrary and reflects a very rough categorisation with partly overlapping meaning depending on the complexity of the substance. There is no unique attribution of substances possible considering the complexity reflected in organic PM.
- Categorisation: The chosen categories rural, urban and kerbside are arbitrary. One could argue as well to take other categories, e.g. industrial, residential, remote. However, the suggested categories reflect a common attribution in LCA and the terminology can easily be coupled with the population densities (city centre corresponds to kerbside; high population density corresponds to urban; low population density corresponds to rural).
- The density of PM is 2 kg/dm³ on average.

Table 12. Most important assumptions and limitations for the background concentration.

8.2 The Fate Model

The aim of the fate model is to link the emissions to a concentration. It contains the dispersion and the consequential dilution of the emission in the atmosphere and the predominant removal processes. Both, transport and dilution depend on particle diameter. Figure 12 shows the relevant removal process depending on particle diameter according to Seinfeld & Pandis¹⁹.

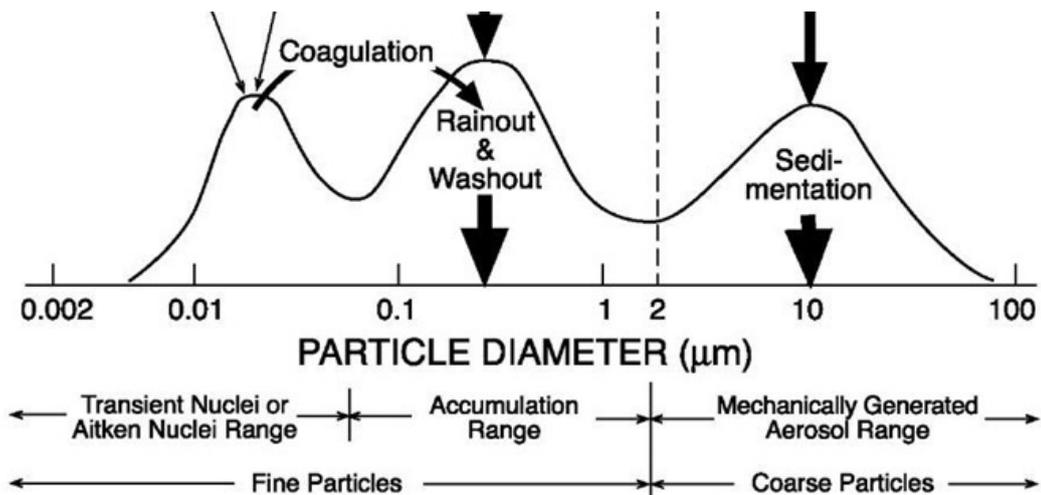


Figure 12. Removal process depending on particle size - Source: Seinfeld & Pandis¹⁹.

The removal processes are calculated as a function of particle diameter on a log scale from 1 nanometre to 10 micrometre (0.001-0.009, 0.01-0.09, 0.1-0.9, 1-10 µm). The result is a unit less fraction of mass or particle number removed.

- dry Deposition (most important for PM_{2.5} – PM₁₀)
- wet Deposition (for UFP – PM_{2.5})
- Coagulation (for UFP)

Dispersion

The dispersion and dilution of the emission depends on several parameters, such as residence time, wind velocity, height of the mixing layer, etc.

Residence time

Residence time of PM depends on the efficiency of the removal process (highly efficient removal process lead to short residence time). Vice versa, the removal of particles depends on the residence time (e.g. longer residence time for dry deposition allows more particles to settle down). Thus, a circular reasoning has to be avoided. This is accomplished by assuming a residence time general valid for all PM size classes. The general residence time is then considered for the calculation of the removal processes. Since the removal process is modelled depending on particle diameter, an overestimated general residence time leads to a small mean concentration for this particle class. Hence, the exposition (expressed as concentration multiplied with the time exposed) accounts for the overestimated residence time assuming underestimated mean exposure compared to the “real” residence time and “real” exposure concentration.

The general residence time was calculated using a formula provided by Wiman¹¹¹ and Jaenicke¹¹²:

$$\frac{1}{\tau} (dr) = \frac{1}{K} * \left(\frac{r}{R}\right)^2 + \frac{1}{K} * \sqrt{\left(\frac{r}{R}\right)} + \frac{1}{\tau_{wet}}$$

With:

- $K = 1.28E+08$ seconds, (empirical value)
- $R = 0.3 \mu\text{m}$, radius at which particles derive the maximum residence time
- r = particle radius
- $\tau_{wet} = 6.90E+05$ s, empirical value for mean residence time for the lowest 1.5 km of the troposphere (8 days). The mean residence time for a generic particle mixture results in 4.45 days.

Travelling distance

The travelling distance for such a particle mounts up to 1538 km using a mean wind velocity of 4 m/s.

Dispersion model

An Eulerian 1Box-Model is used for the dilution of the emissions (assumed to be monodisperse) in the atmosphere with a mean travel distance of about 1540 km and a mean residence time of 4.5 days. The height of the mixing layer is assumed to be 800 m. Derwent et al.¹¹³ and Metcalf et al.¹¹⁴ applied a fixed height of 800 m for the mixing layer. This was considered as a reasonable average over all conditions by Jones¹¹⁵. These models proved to provide modelled concentrations close to measured data. 800 m are therefore taken as a reasonable best estimate for the mixing height in Europe. Hence, an emission is diluted into a cube with a length of 1540 km, a height of 0.8 km and a width of 1 km. The width of the model has no influence on the results on exposure. An alternative model with a larger width there is more dilution (smaller exposure concentration) which affects more people, whereas in a model which is less width the dilution is smaller (higher exposure concentration) and in return less people are affected. In short, combining the fate and the exposure model results in a fractional arithmetic in which the model width can be reduced.

After 1500 km or 4.5 days it is estimated that, on average geographical conditions, either the ocean or the high-Alpine area limits exposure and the successive uptake of PM. This value could be over- or underestimated, however, for a generic model as it is used in LCA this is an appropriate assumption.

Several studies proved that the height of the emission (ground level by a mobile source, or a high stack from an industrial process) has high impact on the dispersion of the emission. Humbert et al.¹¹⁶ summarises these results and suggests intake fractions depending on stack height. From this work dilution factors are derived and it is accounted for three different stack heights:

- ground level (1 m above ground): mobile sources, traffic
- low stack: at 20 m above ground representing typically residential heating
- high stack: at 100 m above ground representing typically industrial process.

The three levels of stack height are coupled with potential emission release for three different site specifications (rural, urban kerbside).

Dry Deposition

The most important parameter for the fraction of dry deposited particles ($f_{dep,dry}$) is the dry deposition velocity (v_d) which depends on the particle diameter d_p . In addition, wind velocity (v_{wind}), the travel distance (x) of the particles and the height of the mixing (H) layer has to be determined.

$$f_{dep,dry} = 1 - \exp\left(-\frac{v_d * x}{H * v_{wind}}\right)$$

Whereas x divided by v_{wind} represents the residence time.

Wet Deposition

The fraction of wet deposited particles depends on the fraction of time when it is raining (F_{rain}), the residence time in the atmosphere (represented in the formula below as ratio of travel distance x and v_{wind}) and the scavenging coefficient.

$$f_{dep,wet} = F_{rain} * \frac{x}{v_{wind}} * \lambda$$

The fraction of time with rainfall is derived from the annual rainfall (A_{rain}) and the rainfall intensity per rainfall event (I_{rain}):

$$F_{rain} = \frac{A_{rain}}{I_{rain}}$$

With:

- A_{rain} = 671 mm on average per year
- I_{rain} = average rainfall intensity of 1.3 mm per hour

The fraction of time with rainfall events averages to 7%.

The precipitation scavenging coefficient (λ) describes the loss of particles from the atmosphere by their incorporation into raindrops. λ depends on the diameter of the raindrop (d_r), the vertical velocity of the raindrop (v_t), the number of raindrops ($N(d_r)$) in a rainfall event and the collision efficiency (E) which is a function of particle and raindrop diameter. E can be interpreted as the probability of the rain droplet to hit a particle when falling down.

$$\lambda = \frac{\pi}{4} * d_r^2 * v_t * E(d_p, d_r) * N(d_r)$$

Raindrop diameter (d_r): 1mm

Vertical velocity of the raindrop: 4 m/s

The number of raindrops per unit volume was calculated from total annual rain fall and assuming monodisperse, spherically shaped raindrops with a diameter of 1mm. The collision efficiency was taken from Andronache¹¹⁷.

Brownian coagulation

Brownian coagulation is a process that reduces efficiently the number concentration of UFPs. Whenever 2 particles coagulate, the total number of particles is reduced by 1¹⁹. But there is a new born particle with a diameter equal to the sum of diameters of both particles. Hence, the change in PNC (N) for a pair of particle diameters due to coagulation can be expressed as:

$$\frac{dN}{dt} = -K_{1-2} * (N(d_{p1}) * N(d_{p2}) + N(d_{p1} + d_{p2}))$$

and,

K_{1-2} : is the Brownian coagulation coefficient depending on both particle diameters.

The Brownian coagulation coefficients for particles between 1 nm and 10 μm are presented in Figure A1 in the Annex. This calculation has been made for any possible pair of particle diameter between 1 nm and 10 μm . The PNC decreases quickly and produces after very short time a new particle distribution through the changes due to coagulation. The initial particle number distribution at $t=0$ was taken over from Anderson⁴⁵. Then, a coagulation step was executed with a time step of 1 second. After this coagulation step, the new background concentration was calculated by using the initial PNC, subtraction the removed number of particles for each particle diameter and adding the new born particles. This procedure was then repeated for a sequence of 55 time steps:

- 1 – 10 sec, per sec.
- 20-100 sec, per 10 sec.
- 200-1`000 sec, per 100 sec.
- 2`000-10`000 sec per 1`000 sec.
- 20`000-100`000 sec per 10`000 sec.
- 20`000-1`000`000 sec, 100`000 sec. (about 10 days)

Results for the combined removal processes

The fraction of mass deposited with respect to all removal processes is presented in Figure 13.

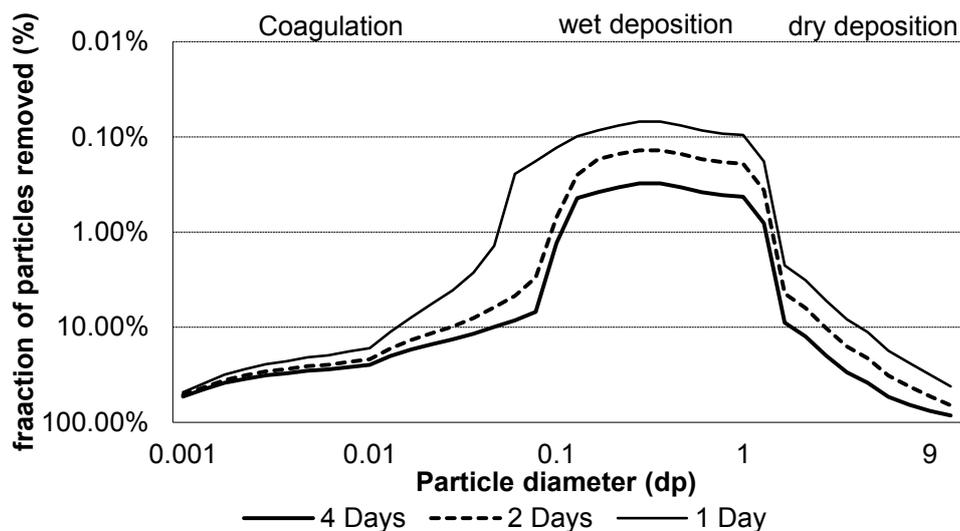


Figure 13. Results removal processes

Figure 13 shows that after a few hours, very small and very large particles are significantly reduced in the atmosphere. However, for particles between 0.1 and 2 μm , nearly the same amount of particles is observed in the air even 4 days after releasing the particles.

Assumptions and limitations for the fate model

- Dispersion: A one-box Euler model is assumed for dispersion of PM in the atmosphere.
- The emission is assumed to follow a monodisperse distribution.
- Height of the mixing layer: The height of this layer is not constant. In this simplified model it is assumed to be at 800 m above ground. The height is an important parameter for the volume in which the emission is dispersed.
- Log scale: A log scale is taken throughout the entire fate model in order to reduce the number of data points: 1nm to 10 μm results in 36 datapoints to be calculated instead of 10`000 datapoints (1 nm step, no log scale)
- For wet deposition the mean values from Jolliet & Hauschild¹¹⁸ have been used with respect to average rainfall intensity (1.3 mm/h,m²) and the mean diameter of a rain droplet (1mm). The annual rainfall of 671 mm is an average calculated from a geographical website¹¹⁹.
- Coagulation: Two important assumptions for the coagulation process should be mentioned here:
 1. Coagulation is modelled assuming all particles to be spherical.
 2. Each collision between 2 particles leads to a coagulated particle with the size being the sum of both particle sizes.
- Coagulation in laminar shear flow, turbulent flow and from gravitational settling is neglected in this model since it is not relevant for UFP.
- There are a number of processes which are not considered in this model but may have relevant impact:

1. Interactions of particles with water vapour in the air, for example, in clouds or fog.
 2. The formation of secondary organic particles.
- The latter two processes are not understood well enough in order to integrate them in an impact assessment model.
-

Table 13. Most important assumptions and limitations for the fate model

8.3 The exposure model

In the exposure analysis the derived concentration from the fate model is linked to a dose. The exposure of a single person is modelled calculating first an intake fraction followed by the calculation of the uptake fraction.

The particle Intake

The intake fraction (f_{intake}) can be described as the mass of particles inhaled due to exposure to ambient air.

$$f_{intake} = C_{exp} * BR * \tau * f_{outdoor}$$

With

- C_{exp} : the exposure concentration of PM (result from the fate model)
- BR: the breathing volume of a single person over one day (13 m^3)¹²⁰
- τ : mean residence time
- $f_{outdoor}$: fraction of time a person spent outdoors^{121, 122}

The particle uptake

Modelling uptake determines the amount of particles deposited in the different respiratory tracts. The respiratory system is split into three different compartments: the naso-pharyngeal, the trachea-bronchial and the alveolar department.

$$Uptake = Intake * f_{uptake}$$

The uptake is the result of the multiplication of the intake calculated above and the fraction of particles deposited in the respiratory system. The particle deposition model of Theophanides et al.¹²³ was applied.

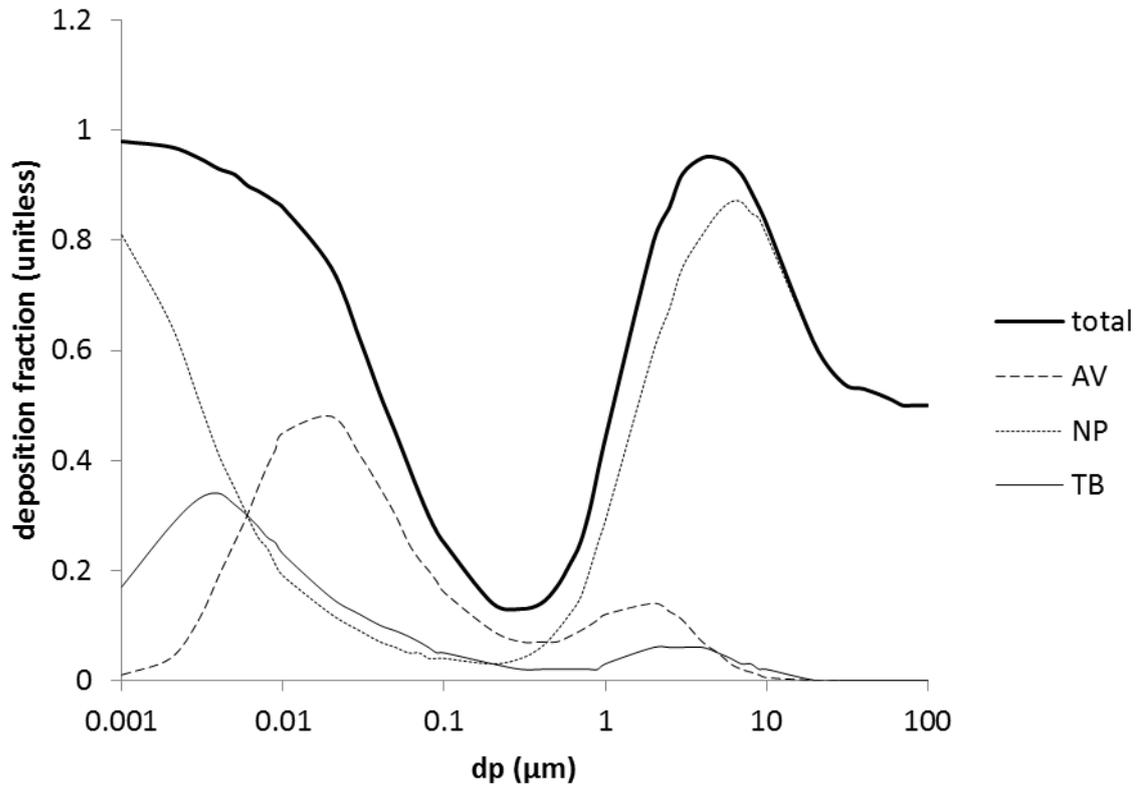


Figure 14. Uptake per respiratory department: AV: alveolar; NP: naso-pharyngeal; TB: trachea-bronchial.

Figure 14 shows the deposition fraction of the 3 different respiratory compartments as a function of particle diameter (dp). The total uptake is the superposition of the uptake of the three respiratory compartments.

Comparing the results of the removal processes from the fate analysis and the total uptake in the respiratory system leads to the important finding that the removal of particles in the range of 0.1-1 μm is very small, but these particles don't tend to stick in the lung tissue but rather are exhaled (Figure 15).

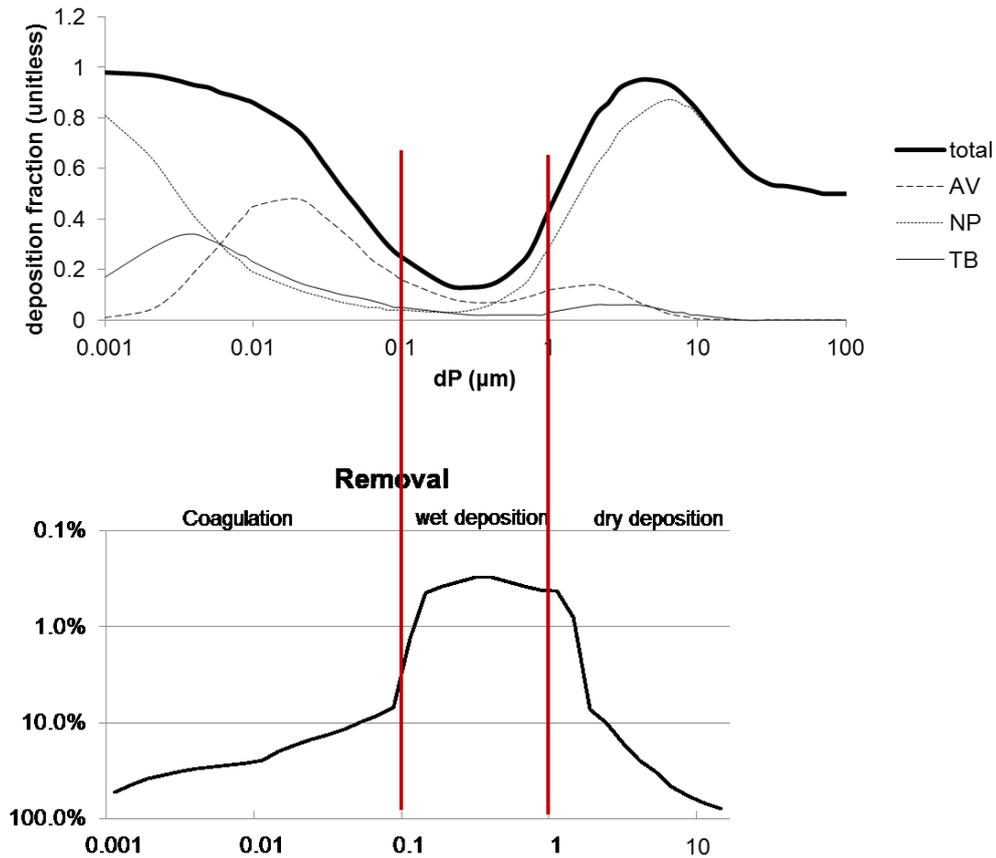


Figure 15. Particle removal and particle deposition as a function of particle diameter.

This finding has major influence on the assessment of the damage on human health of particles when it is expressed per kilogram. Furthermore, the finding puts the potential damage of particle in the range between 0.1 and 1 μm in perspective.

Population uptake

The result from the uptake of PM of a single person has to be expanded on to the total affected population. Thus, the uptake of a single person is multiplied by the population density.

In ecoinvent air emissions are recorded as emitted to an unspecified, a low population density or high population density site, without a clear distinction of these three categories¹²⁴. However, this indication is meaningful for the outcome of human health impacts. The model accounts for 3 different population densities:

City centre population density: 5000 P/km²

High population density: 500 P/km²

Low population density: 50 P/km².

It is assumed that an “unspecified” population density accounts for an equal population density as high population density. Thus, it is not mandatory to keep this term separated. Disregard the category “unspecified” helps to reduce the final set of CF.

Note that the width of the Eulerian 1-Box is arbitrary and does not influence the population uptake because the model considers:

- A monodisperse distribution of the emission in the Box, and
- A monotonic population density.

It follows that, doubling the width of the model results in a doubling of the number of people affected but in a bisection of the concentration of the emission in the air. However, the population uptake remains the same.

Assumptions and limitations for the exposure model

- The model for the uptake assumes all particles to be of spherical shape
 - Some restriction of this model arise due to the fact that no further particle characteristics like shape, van der Waals force or electric interaction with tissue in the respiratory system were considered. Solubility of particles is considered in the effect modelling.
 - Although the total deposition of particle has been modelled per respiratory compartment, the particle deposition is not split into the different compartments, since there is a missing link between the type of particle which causes a specific disease per lung compartment. Hence, total uptake is considered when linking exposure to human health effects.
 - Ambient air exchange per person is assumed to be 13 m³/day. This reflects a breathing volume of an average person under average activity level¹²⁰.
 - For the population density it is assumed that the population is monotonic distributed. This is in line with the monodisperse distribution of the emissions and leads to the fact that the the result gets independent of the width of the model.
-

Table 14. Most important assumptions and limitations for the exposure model.

8.4 The effect model

The particles deposited in the respiratory system represent a dose. This dose is linked to human health effects. In chapter 3 the physical properties “size”, “chemical compositions” and “solubility” were considered as suitable properties for the purpose of this study.

The effects of size

The size effects are considered using the following size categories and the corresponding measure:

- Ultrafine Particles (<100 nm): measuring number concentration
- Fine particles (100 nm - 2.5 µm): measuring mass
- Coarse particles (2.5 µm - 10 µm): measuring mass

The results derived in the literature study for size are used for the assessment of the effects of size. The results are expressed as relative risk (RR) or odds ratio (OR) and presented in Figure 16.

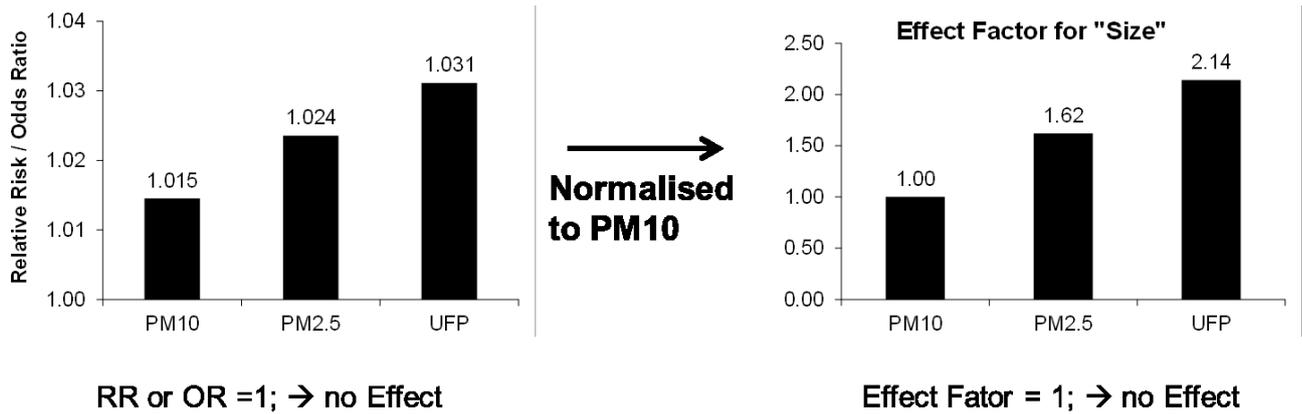


Figure 16: Relative risk and Odds ratio (left panel) and after normalisation to PM₁₀ (right panel)

The literature study on size effects of PM yielded in a risk for effects on human health of 1.5% for PM₁₀, 2.4% for PM_{2.5} and 3.1% for UFPs. After normalisation to PM₁₀ this translates into unit less effects of size for PM₁₀ being equal 1, for PM_{2.5} equal 1.62 and for the UFP equal 2.14. This result is interpreted as follows: UFP in the ambient air cause 2.14 times more damage than PM₁₀, and PM_{2.5} cause 1.62 times more damage to human health than PM₁₀. This result is already independent of a given increase in of the pollution level. It accounts for the relation between the different particle size classes.

The effects of chemical composition

The results from literature study accounting for the damage to human health due to chemical composition can't be used because of the weak data basis. Useful data were found in a US database which provides unit risk factors (URF) for lung cancer disease and reference concentrations (RFC) for cardiopulmonary disease⁹⁰. The comprehensive database contains URF and RFC of about 240 chemicals found in ambient air (Table A2 in the Annex). The database describe a useful application for their data as following: "These unit risk factors and reference concentrations can be used as health benchmarks, to evaluate the potential health effects of air toxic concentrations."⁹⁰ This is in line with the way the data are in fact used in the effect model.

The chemical components of PM measured in European ambient air (Table A3 in the Annex) allows a selection of suited URF's and RFC's. Base on this list of 240 chemicals the following list of substances were taken over (Table 15) for the chemical characterisation of PM in ambient air:

inorganics, non-metals	metals	organics
1 Carbon Black	9 Arsenic (inorganic)	22 Alkanes/Alkenes/Alkines
2 Nitrate	10 Barium	23 Aldehydes
3 Ammonia	11 Cadmium	24 Ketones
4 Sulfates	12 Chromium VI	25 Alkanol
5 Inorganic salt	13 Cobalt	26 Organic Acid
6 Selenium and compounds	14 Copper	27 Aromatics (monocyclic and polycyclic with heteroatoms)
7 Mineral dust	15 Iron	28 PAH
8 Asbestos	16 Lead	29 PCB
	17 Manganese	30 Dioxins, PCDD
	18 Mercury (elemental)	31 Furane, PCDF
	19 Nickel and compounds	32 Organic matter
	20 Vanadium or vanadium pentoxide	33 Organic matter without PCB, PAH, dioxin/furan
	21 Zinc	34 unspecified chemistry

Table 15. List of chemical substances and groups of substances used for the chemical description of PM.

Table 15 covers the most important substances found in ambient air for these substances for which a corresponding toxicity indicator is available. A couple of substances should serve as proxy (e.g. organic matter or unspecified chemistry). This allows that inventories don't need to provide full chemical disclosure of PM emissions. This impact assessment method allows for this by providing different chemical groups of substances. For example, dioxin, furan and PAH can be assessed separately or within organic matter using "Organic matter, total". If the substances are assessed separately, either all organic components are disclosed and assessed separately (alkanes, ketones, aldehydes etc.) or one choses "Organic matter without PCB, PAH, Dioxin and Furan". In a worst case, PM is not chemically disclosed at all. Then the applicator choses "unspecified chemical composition".

Few substances (carbon black, iron, zinc) were not listed in the database. Since these substances contribute significantly to the ambient air PM mass, they might contribute to human health effects substantially. Hence, the literature survey is used for chemical substances to find a proxy substance with similar RR or OR as the one missing in the database and accordingly the missing substance got the same URF or RFC as the substance known from the database. The advantage using the literature search instead another toxicology database is that the data from the literature search provides the direct information to human health effects. Using another toxicology database would probably generate a consistency problem since we do not know how toxicology indicator from each database was elaborated and whether they fit together. In order to find a suitable proxy for a missing substance in the toxicology database the following restrictions were applied:

- The number of paired data has to be bigger than 8 (→ statistics possible).
- The P-value has to be bigger than 0.1.
- The absolute RR difference between the proxy substance and the missing substance has to be smaller 0.005 (=0.5%).

For example: Zinc is not listed in the toxicity database. The literature search provides a comparison between Copper and Zinc. The number of paired samples is 14 and the P-value from the paired t-test is P=0.79. The difference in RR between copper and zinc is 0.001 (=0.1%). Consequential, copper is chosen as a proxy for zinc and the URF and RFC values from copper are assigned to zinc. This procedure was applied only for substances known to be low toxic.

The effect of the single chemical was derived as a multiplication of the ambient air concentration of a specific substance multiplied with the URF (for lung cancer) and the RFC (for cardiopulmonary disease). The total effects of chemical substances for lung cancer were calculated the following:

$$E_{Chemistry} = \sum_{substance\ i}^{substance\ n} URF_i * ambient\ air\ concentration_i$$

The total effects for CVD were calculated similar using RFC instead of URF. The total effect of the chemical substances is calculated as the sum over the effects of the single chemicals.

The effects of solubility

For an assessment of solubility information is used from:

- the effects of size,
- the effects of chemical composition, and
- The effects of solubility of the PM emission

With regard to soluble Particles, it is assumed that these particles resolve fast. Since they disappear quickly they lose the effects due to size – no matter whether UFP, PM_{2.5} or PM₁₀. Further, we account for the full potential toxic effects of all substances attached to the particles or hidden within the particle due to the fact that the particle resolves and all the toxic substances are released (Table 16).

	soluble	non soluble
PM10: size	E=1	E=1
PM10: Chemistry	E= ∑ E of chemicals	E= ~ to ratio Surface/Volume
PM2.5: Size	E=1	E=1.62
PM2.5: Chemistry	E= ∑ E of chemicals	E= ~ to ratio Surface/Volume
UFP: Size	E=1	E=2.14
UFP: Chemistry	E= ∑ E of chemicals	E= ~ to ratio Surface/Volume

Table 16. Modelling of solubility depending on size and chemistry.

Non soluble particles keep the size effect derived from the literature study. However, the influence of attached or hidden chemicals within a particle is diminished due to the fact that certain substances within insoluble particles get never/reduced into contact with human tissue. This effect is proportional to the ratio Surface/Volume of a spherical particle (all particles are assumed to be spherical).

$$E_{solubility} = \frac{particle\ surface}{particle\ volume} = \frac{\pi * dp^2}{\frac{1}{6} * \pi * dp^3} = \frac{6}{dp}$$

Solubility is implemented for 3 different levels of solubility.

- soluble (75% of the particles are soluble, 25% are non-soluble)
- non-soluble (75% of the particles are non-soluble, 25% are soluble)
- unspecified solubility (50% of the particles are soluble, 50% are non-soluble)

Applying the ratio surface/volume for different particle sizes leads to the following effects for solubility depending on the three levels of solubility (Table 17):

	insoluble	soluble	unspecific
level of solubility	25%	75%	50%
PM ₁₀	0.75	0.25	0.50
PM _{2.5}	0.48	0.20	0.34
UFP	0.60	0.87	0.73

Table 17. Effects of solubility depending on particle size.

Assumptions and limitations for the effect model

Effects of size

- Particles bigger than 10 µm are neglected. This is justified by the rather short residence time of these particles and the fact that these particles stick to the naso-pharyngeal tract (Figure 14). Hence, they do not get into contact with tissue sensitive to CVD or lung cancer.
- The results from the epidemiological studies for UFP are not solely based on long term studies. This leads to higher uncertainty on the effects on human health.
- Although OR and RR are not exactly the same, the analysis of the results from the epidemiological literature search does not differentiate between the two measures. This is a suitable procedure as long as the values of RR and OR do not differ too much^{30, 31}.
- As a limitation for the effects of size one could argue that two different techniques for measuring the amount of particles have to be applied. While for UFP the PNC has to be determined, for PM₁₀ and PM_{2.5} the mass shall be measured.

Effects of chemical composition

- The chemical composition is the most sensitive property influencing the final outcome of the damage to human health. The toxicity values are taken from a toxicity database. Since the values don't stem from epidemiological studies the real effect from the system physiology point of view underlines potentially higher uncertainties compared to size effects. However, the data are not used to derive directly cases for lung cancer and CVD, but rather as benchmark values in order to allocate the suitable proportion of health effect to each substance.
 - Few substances with very low toxicity but a significant share in the ambient PM are not listed in the toxicology database. The toxicity values for these substances are approximated based on the effects of the epidemiological literature study for chemical substances.
-

Effects of solubility

- The model for the effects of solubility assumes all particles to be of spherical shape
- The model for the effects of solubility assumes all particles to have a given density (2 kg/dm³) and according to this shape a defined volume.
- If a particle is considered to be soluble it is assumed that it dissolves very fast (so fast that it is completely dissolved before the metabolism disposes the particle).

Table 18. Most important assumptions and limitations for the effect model.

8.5 The damage model

The human health effects derived from the effect modelling have to be translated into damage to human health. The total damage caused by air pollution is already known and quantified for Europe from a study of Lopez¹²⁵. Table 19 presents the total DALY, DALY due to lung cancer and DALY due to CPD disease for Europe.

1418

Comparative Quantification of Health Risks

Table 17.12 Attributable YLL and DALYs for cardiopulmonary disease, lung cancer, ARI^a and total mortality

Subregion	Cardiopulmonary disease		Lung cancer		Acute respiratory infections		Total		% change
	YLL (000s)	DALYs (000s)	YLL (000s)	DALYs (000s)	YLL (000s)	DALYs (000s)	YLL (000s)	DALYs (000s)	
AFR-D	162	193	4	4	119	121	285	319	12
AFR-E	84	100	3	3	61	62	147	166	13
AMR-A	116	161	37	38	0	0	152	200	32
AMR-B	201	273	20	20	11	14	232	307	32
AMR-D	31	39	1	2	11	12	44	53	21
EMR-B	65	77	5	5	7	9	77	91	18
EMR-D	386	457	17	17	155	162	558	636	14
EUR-A	90	122	27	28	0	0	117	151	29
EUR-B	238	286	30	31	20	21	288	338	17
EUR-C	291	340	27	28	2	2	320	360	13
SEAR-B	240	291	22	22	21	25	282	339	20
SEAR-D	1 006	1 195	56	57	250	261	1 312	1 513	15
WPR-A	65	95	18	18	0	0	84	114	36
WPR-B	1 992	2 732	304	317	204	224	2 504	3 272	31
World	4 966	6 360	572	591	862	913	6 404	7 865	23

^a In children aged 0–4 years.

Table 19. DALY's for EUR-A countries provided by Lopez et al.¹²⁵.

The 28'000 DALY for lung cancer are allocated to the effect factors found for lung cancer in average ambient air in Europe according to their weight and the 122'000 DALY for cardiopulmonary disease are allocated to the effect factors found for cardiopulmonary disease.

Assumption and Limitations for the damage model

- No assumptions had to be taken for the damage model.
-

Table 20. Most important assumptions and limitations for the effect model.

9 The characterisation factor (CF)

The CF can now be calculated by combining the models for Fate, Exposure, Effect and Damage and considering the results derived from the characterisation of PM in ambient air. The CF for a substance i is expressed as:

$$CF_i = \frac{Em_i * F_i * Ex_i * Ef_i(\text{size, chemistry, solubility}) * D_i}{\text{amount of substance}_i \text{ in ambient air}}$$

- The units referring to the formula of the CF are: Emission (Em): kg
- Fate (F): $1/m^3$ (from dilution; removal are unit less fractions)
- Exposure (Ex): m^3 (from air inhaled; uptake is modelled as unit less fraction)
- Effect (Ef): risk/kg (from chemistry; size and solubility are unit less numbers)
- Damage (D): in DALY due to PM per “risk effect”
- Amount of substance: in kg or number of particles

Out of this multiplication the CF gets the unit DALY per kg of a substance.

This computation has to be done for all substances separately for lung cancer and cardiopulmonary disease. The total CF is then sum of the CF lung cancer and the CF CPD.

9.1 Base model

Since the DALY in Table 19 represents the damage due to air pollution in Europe, the CF have to represent average conditions in Europe with respect air pollution concentration and affected population. Thus, from the choice of all possible parameters considered for fate (stack heights, emission site), exposure (population densities) and effects (size, solubility) a base case was defined in a way to represent best European conditions. The base case has the following characteristics:

- Stack height: low (20 m above ground)
- Emission site: urban
- Population density: high (500 P/km²)
- Solubility: unspecified (50% of particles soluble, 50% non-soluble)

The DALY's were allocated to the parameters according to this base case. The number of CF representing the base case is 93. The number is made up of 3 size classes (PM₁₀, PM_{2.5}, UFP) each containing 31 chemicals (CB, PAH, PCB, Cadmium, etc.) and belong to the base case.

Overall, 2781 CF's were calculated. The number is build up by the 93 CF from the base case have to be multiplied with the number of different settings possible due to changes of the base case scenario (solubility: 3; population density: 3; stack height: 3) plus some CF calculated in case of unspecified chemical resolution of PM (270 CF's).

The CFs have to be tested and verified. The verification of the model is done by analysing the degree to which the model assesses what it is supposed to measure. The CFs for the base model are listed in the Annex (Table A4).

10 Verification of the Model

A first check of the model can be achieved by analysing the CF. The model works correct, if the sum of all CF over PM₁₀ compared to the sum of all CF over PM_{2.5} or UFP reflects the weights we derived from the literature study (see chapter 7.4). In fact, this step represents an artificial release of 1 kg PM for every substance with a CF in this model. The same idea is applied in order to verify solubility, population density and stack height.

10.1 Size and solubility

Figure 17 shows the weight of the CFs split into size and solubility. Non soluble particles have a weight of 28 %, particles with unspecified solubility 33% and soluble Particles 39%. This result precisely reproduces the weights derived for solubility in chapter 7.4. Hence, it is confirmed that the impact assessment model assesses solubility correctly according to the idea implemented in the effect model. Further PM₁₀ (3.6%+10.8%+7.2%=21.6%) compared to PM_{2.5} (6.7%+15.9%+11.3%=33.9%) and UFP (17.6%+12.2%+14.9%=44.7%) results in a ratio of UFP: PM_{2.5}: PM₁₀=2.1:1.6:1. Thus, this result is also in perfect accordance with the values used for the effect model of size (see chapter 7.4).

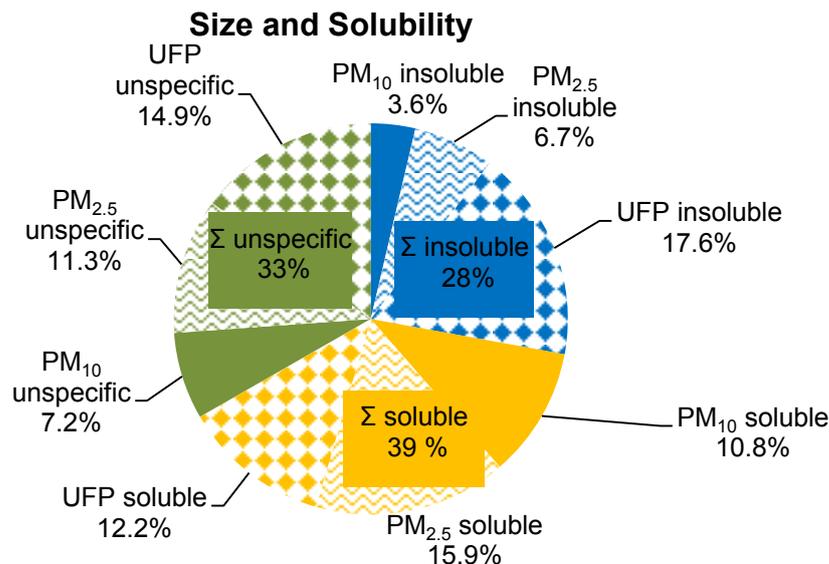


Figure 17. Model verification for size and solubility.

10.2 Population density

The population densities selected in the model are:

- City centre population density: 5000 P/km²
- High population density: 500 P/km²
- Low population density: 50 P/km²

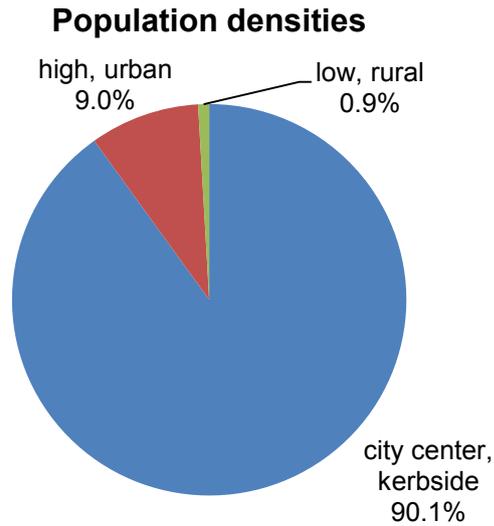


Figure 18. Model verification for population densities.

The ratio of the population densities (5000:500:50) meets the percentage ratio calculated as the sum of all CF with the same population density (Figure 18; 90.1:9.01:0.901) exactly.

10.3 Stack height

The dilution ratio of the PM concentration due to different stack heights is:

- Ground level: 1 (no dilution)
- Low stack: 0.34
- High stack: 0.25

This ratios 1:0.34:0.25 expressed in % results in 62%:22%:16%. Figure 19 depicts that also the model outcome for the stack heights passes the verification test.

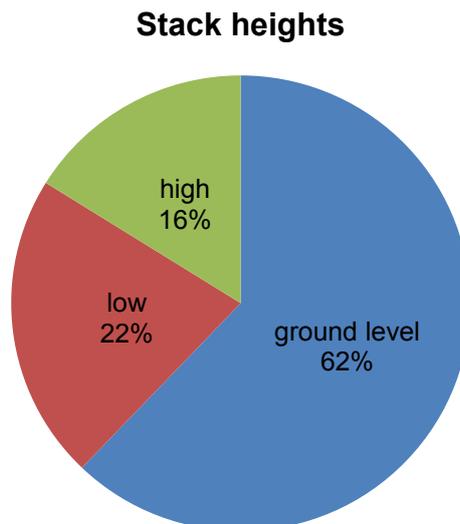


Figure 19. Model verification stack heights.

10.4 Chemical substances

Chemical substances can't be as easily compared as the other properties due to two main reasons. First, the chemical composition of ambient air is not the same for the three size classes. Second, the CF depends on the URF and the RFC. Splitting the CF, based on URF and RFC in its components and calculating ratios between the chemical substances could be done, but only with huge effort. However, such an effort would rise above a reasonable effort. Despite it is worth to have a short look at the CF of the different chemical substances in the base case scenario.

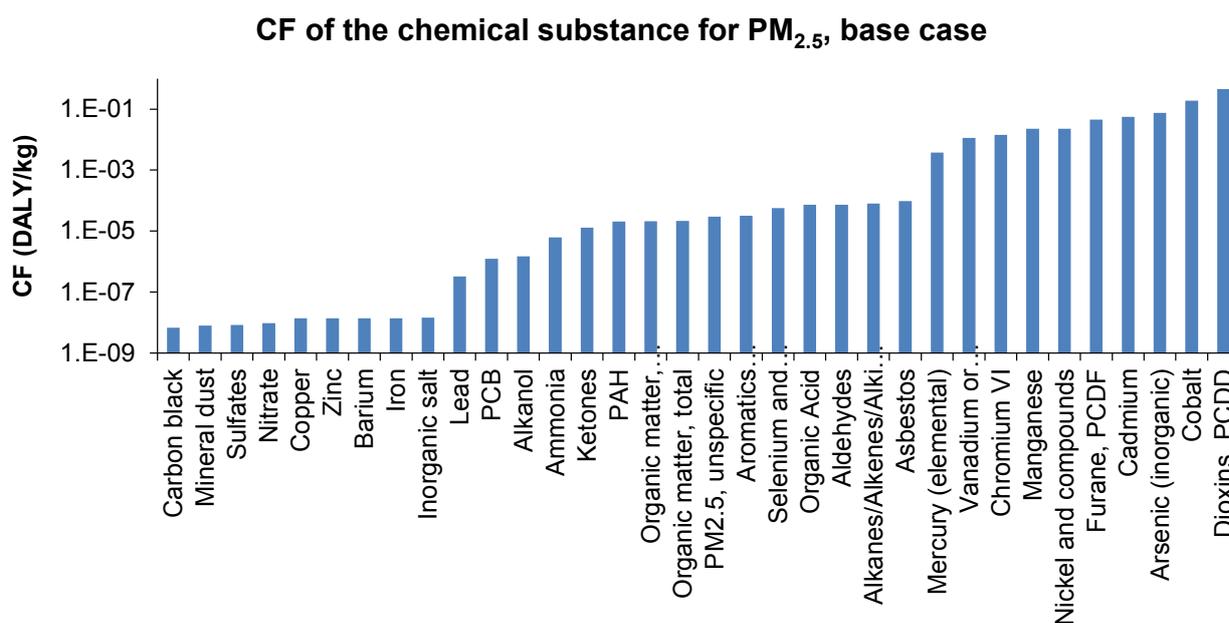


Figure 20. CF for all chemicals for PM_{2.5} for the base case.

Figure 20 presents the CF in DALY per kg PM_{2.5} emitted. Dioxin has the highest damage potential per kg of a substance. The value is about 8 orders of magnitude higher than the values for carbon black, mineral dust or nitrates. The high value for cobalt compared to dioxin is rather surprising since the URF for dioxin is three orders of magnitude higher than the URF for cobalt. The two main reasons for this are:

- Dioxin has a high URF, but there is no effect on CPD. Cobalt has a lower URF than dioxin, but it has also impact on CPD.
- The DALYs for CPD (122'000 DALY) have more weight than the DALY for lung cancer (28'000 DALY).

10.5 Allocation of damage to human health by chemical components of PM in ambient air

Another, more applied way to relate chemical substances among each other provides the assessment of the chemical components of ambient air. This allocation is presented separately for lung cancer (Figure 21), cardiopulmonary disease (Figure 22) and the total damage (Figure 23).

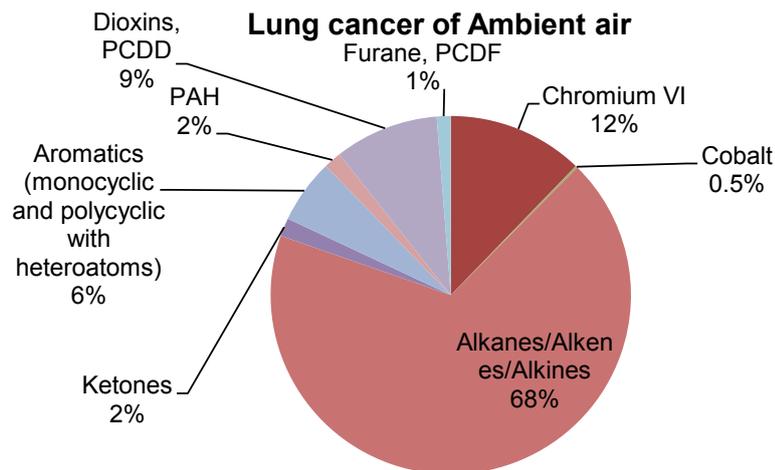


Figure 21. Damage of lung cancer split into the main PM air pollutants

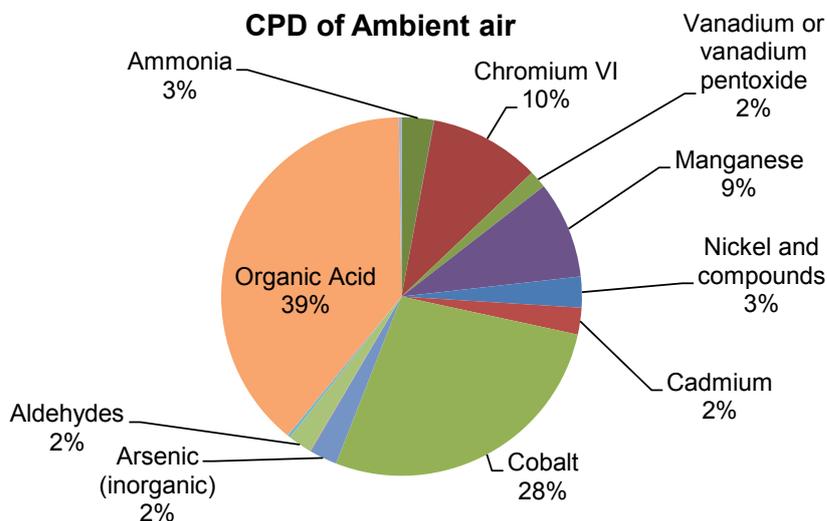


Figure 22. Damage of CPD split into the main PM air pollutants.

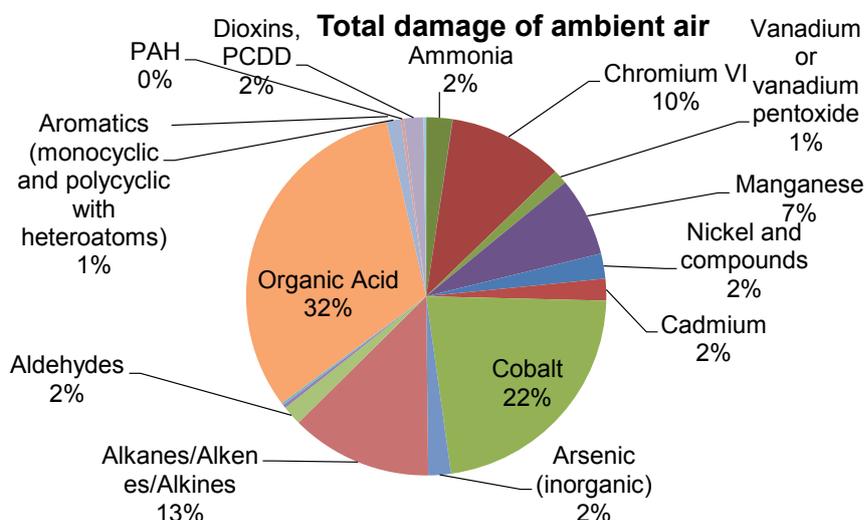


Figure 23. Total damage split into the main PM air pollutants.

Aliphatic and aromatic organic substances contribute significantly to lung cancer. But also dioxins, furans or PAH have rather high shares. From the metals, only chromium and cobalt (0.5%) have a remarkable damage potential.

This looks different with respect to damage to cardiopulmonary diseases. For CPD, metals contribute significantly, but also organic acid and aldehydes have rather high shares on the damage. Among the inorganic and non-metals, only ammonia contributes significantly to CPD.

The total damage is the sum of the damage of lung cancer and CPD. Interesting is the fact that dioxins have a rather small share on the total damage to human health. The share of hexavalent Cr is 5 times higher than the share of dioxin. How is this explained although the URF of dioxin is 3000 times higher than the URF of CrVI?

Three main reasons explain this result: First, the mass of hexavalent chromium is much higher than the mass of dioxins (about 300 times higher). Second, CrVI leads not only to lung cancer as dioxin, but also to cardiopulmonary diseases. And third, according to the study of Lopez the damage due to CPD is more than 4 times higher than the damage due to lung cancer. This last reason explains why other metals, even though they do not appear in Figure 21 (lung cancer), have almost the same shares in Figure 22 (CPD) and Figure 23 (total damage).

From the verification of the CF, the ambient air and the allocation of damage to human health by chemical components it can be concluded that the model does not have (severe) errors. The CF represents the chemical and physical properties exactly the way it is intended. Since it is proven that the model works with respect to solubility, stack height, population density and size, it can be expected that also the effect of the chemical substances are incorporated correctly. This alone does not provide any information whether the model makes sense, but at least the outcome reflects properly the inputs into the model. The quality of the model has to be tested with case studies.

11 Case studies

Five small case studies were accomplished to test the concept of the impact assessment model. The goal of the case studies is

- to show the possibilities the new impact assessment method provides.
- to further validate the model based on case studies (in a less narrow context, using real data instead of applying the CFs only)
- to compare first results with other impact assessment methods

The modelling of the life cycle inventories is not a part of this project, thus the models are rather rough. Hence, the results of the case studies are not suited for further use.

Case study 1: Wood combustion (PM_{2.5})

Case study 2: Wood stoves (PM₁₀ and PM_{2.5})

Case study 3: Small scale wood combustion systems (PM₁₀, PM_{2.5} and UFP)

Case study 4: Sawing natural stoned (mineral dust, PM_{2.5})

Case study 5: heat from pellets and light fuel oil (PM₁₀, PM_{2.5} and UFP)

Aim of these case studies is to further validate the applicability of the impact assessment and to demonstrate the possibilities the model offers. For that reason, the case studies do not represent technical processes that are state of the art, but rather they serve as theoretical models to showcase hypothetical results. The results from the 5 case studies should neither be compared within the case studies nor with other results from the available literature.

11.1 Case study 1: Residential wood combustion of different types of wood

The first case deals with the chemical characterization of fine particle emissions from the wood stove combustion of 5 different types of wood. A series of source tests were conducted on the burning of the most prevalent U.S. tree species in a wood stove in order to characterize the fine particle emissions from wood stoves. Analysis of the wood smoke includes fine particle mass emission factors, organic and elemental carbon content, ionic and elemental composition, and detailed organic speciation.

The study provides a detailed inventory of the chemicals, especially of the organic components. Unfortunately, the inventory is not size-segregated. The inventory refers to PM_{2.5} only.

The emissions were assumed to be of unspecified solubility, emitted in an urban area with high population density out of a low stack. As functional unit 1 kWh heating value of the specific type of wood was chosen and the unit of the damage is therefore expressed as DALY/kWh.

Table 21 provides the summarised inventory of the emission profile compiled from Fine et al.¹²⁶ expressed in kg emissions per heating value of the specific type of wood.

kg Emission/kWh	Typ 1	Typ 2	Typ 3	Typ 4	Typ 5
Carbon Black	7.14E-05	4.27E-05	3.31E-05	1.03E-04	2.15E-05
Nitrate	2.29E-06	4.92E-06	2.45E-06	1.46E-06	1.19E-06
Sulfates	1.72E-06	1.26E-05	1.88E-06	1.39E-06	1.05E-06
Ammonia	7.52E-07	6.47E-07	7.98E-07	2.08E-06	8.49E-08
Copper	3.13E-09	1.29E-08	5.70E-09	7.71E-09	4.53E-09
Manganese	6.27E-09	1.29E-08	1.14E-08	7.71E-09	7.36E-09
Lead	9.40E-09	1.68E-08	5.70E-08	1.54E-08	
Alkanes/Alkenes/Alkines	6.05E-07	5.82E-06	2.57E-06	6.94E-07	6.39E-07
Aldehydes	6.66E-08	1.03E-06	1.57E-07		
Alkanol	1.65E-04	5.27E-04	2.54E-04	2.86E-04	2.03E-04
Acids	6.88E-06	2.83E-05	8.69E-06	2.43E-05	3.41E-06
Aromatics (monocyclic an	1.06E-05	1.40E-04	1.94E-05	1.49E-05	8.39E-06
PAH	1.70E-06	2.79E-06	1.94E-06	3.95E-06	1.74E-06
Furane, PCDF	1.30E-06	1.21E-05	4.88E-06	5.77E-06	2.63E-06
Total	2.62E-04	7.78E-04	3.30E-04	4.44E-04	2.44E-04

Table 21. Life cycle inventory for wood combustion using 5 different types of wood.

The results are presented in the Table 22. 98% of the damage is caused by the furans. Other substances contributing to less than 2% are aromatic and aliphatic organics and manganese. All other substances can be neglected.

DALY	Typ 1	Typ 2	Typ 3	Typ 4	Typ 5
Carbon Black	4.82E-13	2.88E-13	2.23E-13	6.96E-13	1.45E-13
Nitrate	2.18E-14	4.68E-14	2.33E-14	1.39E-14	1.13E-14
Sulfates	1.42E-14	1.04E-13	1.55E-14	1.14E-14	8.64E-15
Ammonia	4.60E-12	3.96E-12	4.89E-12	1.27E-11	5.20E-13
Copper	4.35E-17	1.80E-16	7.91E-17	1.07E-16	6.29E-17
Manganese	1.41E-10	2.91E-10	2.56E-10	1.73E-10	1.66E-10
Lead	3.07E-15	5.51E-15	1.87E-14	5.04E-15	
Alkanes/Alkenes/Alkines	4.84E-11	4.65E-10	2.05E-10	5.54E-11	5.11E-11
Aldehydes	4.80E-12	7.46E-11	1.13E-11		
Alkanol	2.46E-10	7.88E-10	3.80E-10	4.28E-10	3.04E-10
Acids	4.97E-10	2.04E-09	6.27E-10	1.75E-09	2.46E-10
Aromatics (monocyclic an	3.40E-10	4.49E-09	6.21E-10	4.79E-10	2.69E-10
PAH	3.49E-11	5.70E-11	3.98E-11	8.09E-11	3.56E-11
Furane, PCDF	5.84E-08	5.46E-07	2.20E-07	2.60E-07	1.18E-07
Total	5.97E-08	5.54E-07	2.22E-07	2.63E-07	1.20E-07

Table 22. Results in DALY/kWh heat per chemical for each type of wood.

Beside the chemical resolution of the damage it is very interesting to compare the new impact assessment method with other methods (Table 23). Two well-known impact assessment methods using DALY to express damage to human health due to PM serve as reference: Ecoindicator 99 (EI99) and ReCiPe.

Damage in DALY/ kWh	Typ 1	Typ 2	Typ 3	Typ 4	Typ 5
EI99	1.86E-07	5.52E-07	2.34E-07	3.15E-07	1.73E-07
ReCiPe	6.82E-08	2.02E-07	8.59E-08	1.15E-07	6.35E-08
New	5.97E-08	5.54E-07	2.22E-07	2.63E-07	1.20E-07
% share					
New/EI99	32%	100%	95%	83%	69%
New/ReCiPe	88%	274%	259%	228%	188%

Table 23. Results compared with Ecoindicator 99 (EI99) and ReCiPe.

The new impact assessment (indicated with “New”) computes damages very near to the other methods mentioned above. The results assessed with the new IA are higher than those assessed with ReCiPe (except for wood combustion type 1) but lower than those assessed with EI99 (except for wood combustion type 2). The deviation between the new and the established IA methods is less than a factor of three (less than 3 times higher impacts compared to ReCiPe; more than one 3rd compared to EI99).

The study reveals an idea about the variability of possible results when different kinds of wood are burned using an impact assessment that differentiates chemical substances compared to impact assessment which does not compare chemical composition of PM. Most substances can be neglected. In terms of validity, this case reveals that the results are within the same orders of magnitude if compared to other EI99 or ReCiPe. More precise, in this case study the results from the new impact assessment are found by the majority between the two other methods.

11.2 Case study 2: two wood-based household heating systems

In the study from Solli et al.¹²⁷, the environmental effects of two wood-based household heating systems were evaluated. The study describes a comparative life cycle assessment of a wood-based heating system using two different types of stove. This case study considers the combustion process only and provides a not size segregated emission inventory for PM_{2.5} and 5 chemical substances. For the calculation of the results the following assumption are taken: a) Solubility is unspecified, b) population density is high, c) stack height is low. Table 24 shows the LCI which refers to 1 kg wood combusted.

kg emission/kg wood combusted	stove 1	stove 2
PM _{2.5}	6.3E-03	4.0E-02
SO ₂	2.0E-04	2.0E-04
NO _x	9.7E-04	9.7E-04
Cadmium	1.0E-07	1.0E-07
PAH	2.5E-08	2.7E-06
Dioxin	1.0E-12	1.0E-12
Total	7.5E-03	4.1E-02

Table 24. Inventory for particulate emissions from Solli et al. for the combustion of 1 kg wood.

The study assumes all emissions for combustion to be <PM_{2.5}. The mass PM_{2.5} is reflected by the CF for “PM_{2.5}, unspecified chemistry”. Table 25 presents the results expressed in DALY/kg wood combusted for the new impact assessment, for ReCiPe and EI99.

DALY/kg wood combusted	stove 1	stove 2
PM _{2.5}	1.9E-07	1.2E-06
SO ₂	1.6E-12	1.6E-12
NO _x	8.7E-12	8.7E-12
Cadmium	5.3E-09	5.3E-09
PAH	4.8E-13	5.2E-11
Dioxin	4.2E-13	4.2E-13
Total	1.9E-07	1.2E-06
EI99	5.3E-06	2.9E-05
ReCiPe	1.9E-06	1.1E-05
% share		
New/EI99	4%	4%
New/ReCIPE	10%	11%

Table 25. Results in DALY/kWh for the combustion process with two different stoves and a comparison between different impact assessment methods.

Almost all damages are caused by the PM_{2.5} mass. All the other substances could be neglected since they contribute to a much smaller extent (at least 1-2 orders of magnitude less).

The ratio of the damage New/EI99 yields in 4% for both stoves, the ratio New/ReCiPe yields 10% (stove 1) and 11% (stove 2). This might be the result of having a very high share of PM mass assigned to be of unspecified chemistry. The ratio reflects very well the ratio derived for the New CF for PM_{2.5} compared to the CF of ReCiPe and EI99 (Ratios CF for PM_{2.5}: New/EI99=4%; New/ReCiPe=11%). This indicates also that all the other substances do not substantially contribute to the damage in this case study. It remains unknown what damage would be assessed in case the mass of PM_{2.5} would have been chemically analysed.

This case study illustrates that the benefits of the new impact assessment method can only be harvested if the LCI for PM is size segregated and chemically analysed. Otherwise there is no difference to the traditional LCIA methods used in EI99 or ReCiPe. In such a case a comparison of the results between the new and the established LCIA methods reflect the value of the CF for PM.

11.3 Case study 3: Small-scale wood combustion with different stoves

In this study from Lamberg et al.¹²⁸ particle samples from 5 different wood combustion systems were analysed by size (PM₁₀, PM_{2.5}, UFP). Thus, the focus in this case study was put on the different size fractions over different combustion situations. The study provides the number and mass distribution of particles for particle sizes from 10 nm to 10 µm.

The inventory of chemical substances was not as detailed as in case study 1. For example, organic matter was provided but not disclosed in its components, dioxins or furans were not mentioned. For that reason the CF "Organic matter, total" was used for the organic components. It was assumed that all size fractions are chemically identical. The chemically not identified mass is assessed using the CF unspecified for all PM sizes.

The emissions were assumed to be of unspecified solubility, emitted in an urban area with high population density out of a low stack. The functional unit is chosen as 1 MJ heat. Thus, the damage is expressed as DALY per MJ heat. In addition, the apportionment of chemical substances is based on PM₁. In this case study it is assumed that all size fractions have identical chemical composition.

The mass for PM₁₀, PM_{2.5} and the number concentration for the UFP provided in Table 26 bases on the article of Lamberg et al.¹²⁸.

Mass (mg/MJ)	Device 1	Device 2	Device 3	Device 4	Device 5	Device 6
PM₁₀	5.28E+02	9.12E+02	4.41E+02	4.30E+02	1.58E+02	2.50E+03
Pm_{2.5}	4.83E+02	6.27E+02	3.88E+02	3.77E+02	1.41E+02	2.34E+03
PM_{0.1}	1.36E+02	9.00E+01	1.03E+02	1.03E+02	5.90E+01	2.21E+02
PM_{10-2.5}	4.50E+01	2.85E+02	5.30E+01	5.30E+01	1.70E+01	1.54E+02
PM_{2.5-0.1}	3.47E+02	5.37E+02	2.85E+02	2.74E+02	8.15E+01	2.12E+03
number concentration (#/MJ)						
UFP	7.48E+14	9.08E+13	2.63E+14	5.45E+14	3.11E+14	4.73E+14

Table 26. Mass fractions for the different size fractions from Lamberg¹²⁸ per mg/MJ heat produced for all combustion devices.

The mass fraction of the chemical substances used for the LCI are provided in Table 27.

Mass fraction	Device 1	Device 2	Device 3	Device 4	Device 5	Device 6
CB	2.95E-01	7.31E-01	5.43E-01	4.73E-01	5.08E-03	3.80E-01
OM	1.35E-01	2.84E-01	6.78E-02	7.10E-02	4.57E-02	4.68E-01
Cd	2.70E-05	2.69E-05	3.49E-05	6.11E-05	8.63E-06	5.56E-06
Co	3.93E-05	8.21E-05	2.71E-05	2.76E-04	5.08E-06	2.92E-07
Cr	2.09E-05	3.88E-07	1.94E-06	1.97E-06	2.03E-05	4.39E-06
Cu	7.25E-05	2.54E-05	3.88E-05	1.08E-04	3.30E-04	2.92E-07
Mn	9.09E-05	6.42E-04	3.68E-05	5.33E-05	1.02E-03	2.92E-07
Ni	9.95E-06	2.39E-05	1.36E-05	1.22E-05	5.08E-06	2.92E-07
Pb	1.47E-04	4.03E-05	1.40E-04	2.17E-04	1.98E-04	1.84E-05
V	1.23E-06	1.64E-06	5.04E-07	4.14E-07	5.08E-06	2.92E-07
As	1.23E-06	1.49E-06	7.17E-07	1.97E-06	2.28E-06	2.92E-07
Fe	2.58E-04	2.39E-03	4.65E-04	3.94E-04	7.11E-05	2.92E-07
Zn	7.74E-03	4.18E-03	1.41E-02	2.56E-02	4.26E-03	2.84E-03
PAH	1.04E-02	1.41E-02	1.03E-02	6.50E-04	2.62E-06	2.50E-02
unspecified	5.51E-01	-3.64E-02	3.64E-01	4.28E-01	9.43E-01	1.24E-01
Total	1.00E+00	1.00E+00	1.00E+00	1.00E+00	1.00E+00	1.00E+00

Table 27. Mass contribution in % of the chemical substances used in the LCI for all combustion devices.

The emission inventory allows for a size segregated and chemically resolved assessment of the damage to human health due to the PM emissions. However, the derivation of the inventory with respect to the assumption that all size fraction are assigned with identical chemistry is rather artificial.

The total damage resolved for all chemicals is shown in Figure 24. Table A5 in the Annex provides numerical results for all size segregated chemically resolved substances. The

device 4 causes more damage to human health than any other combustion system followed by device 1 and 6.

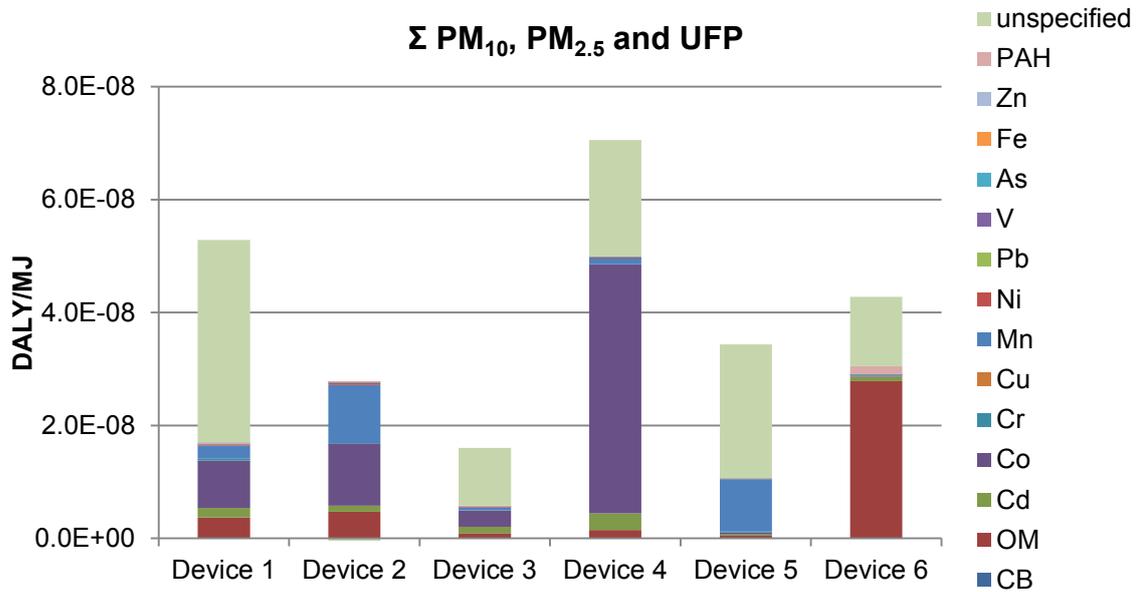


Figure 24. Damage accounted for all size fractions resolved in chemical components.

This can be explained when looking at the UFP fraction. Although the PM mass of the device 4 is small compared to most other systems, the number concentration of particles released is the highest of all system, except for device 1 which has the second worst environmental performance. The difference between the device 4 and 1 is caused by the high cobalt emissions in the ultrafine fraction of device 4.

The size segregated damage resolved for all chemicals for is shown in the Figures 25 - 27.

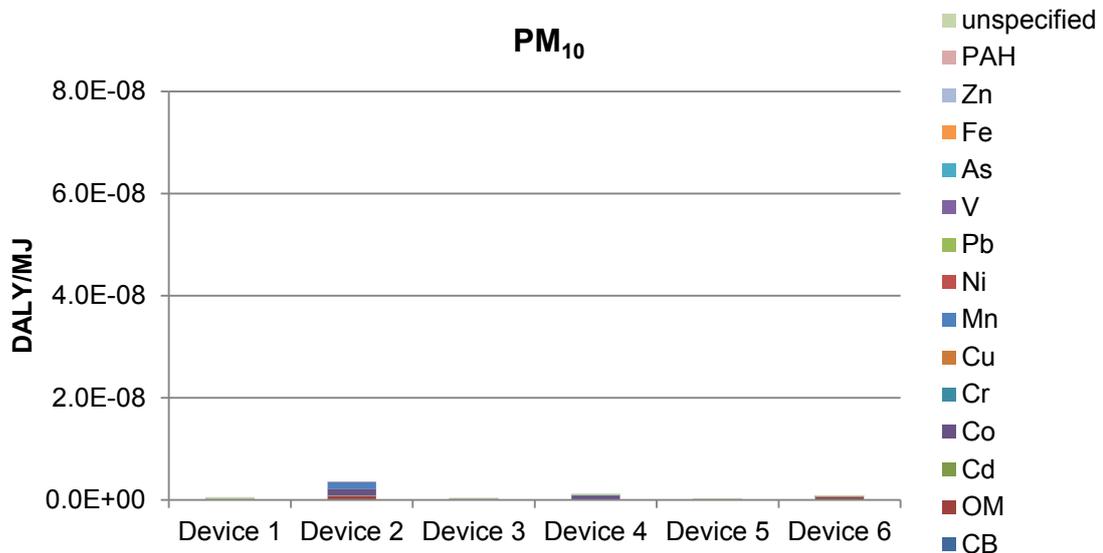


Figure 25. Damage accounted for PM10 resolved in the chemical components

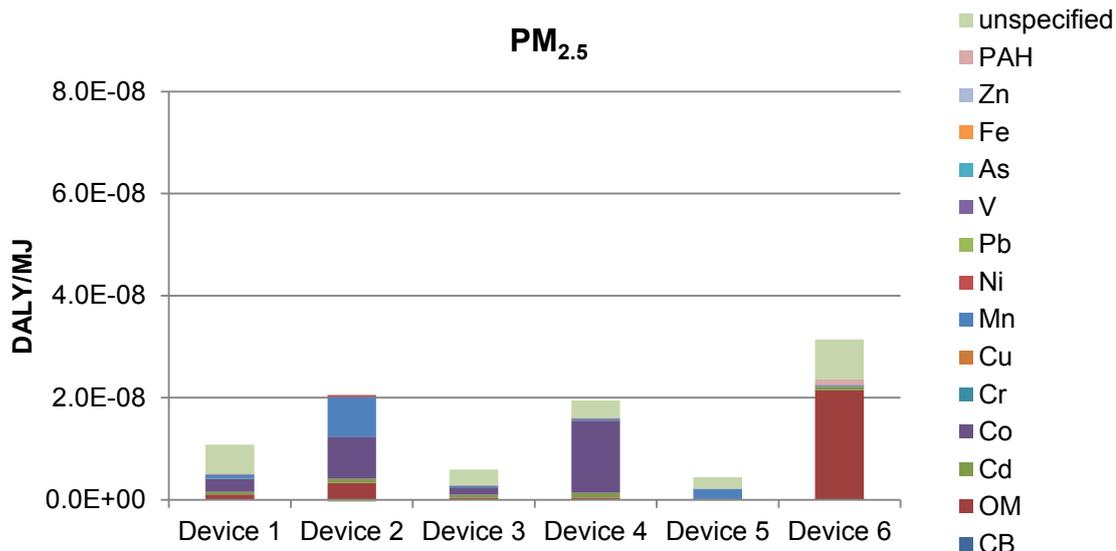


Figure 26. Damage accounted for PM_{2.5} resolved in the chemical components

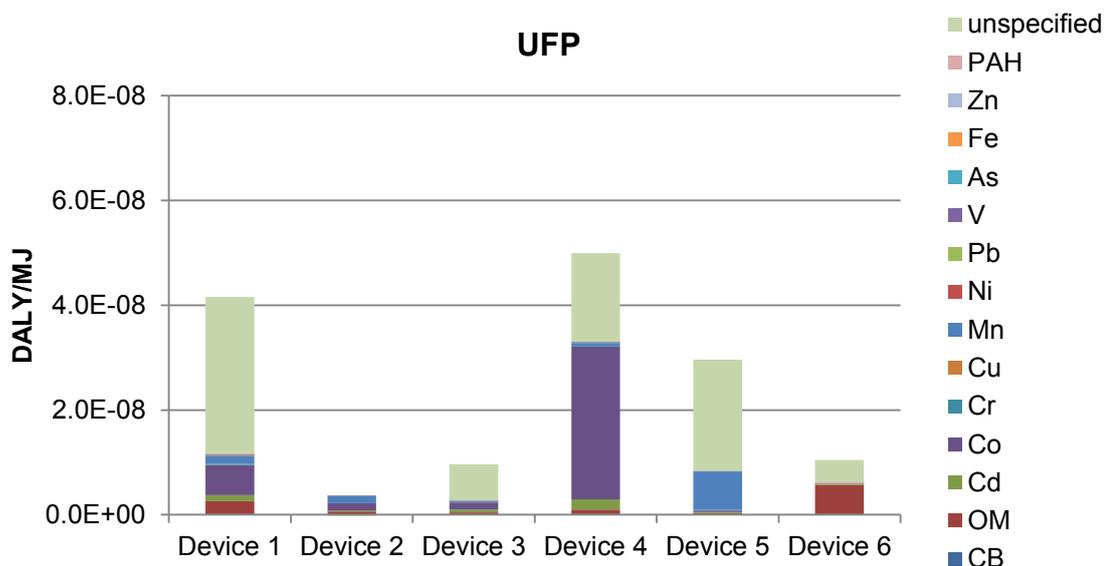


Figure 27. Damage accounted for UFP resolved in the chemical components

From the study setup it is not clear why the device 4 has such a high share of cobalt in the UFP fraction. The fuel was the same for the devices 1 to 4 and device 6 (Birch wood). Commercial wood pellets were used in the device 5. The primary pellet raw material was pine. In the article the chemical composition of the birch wood was analysed. Cobalt is not listed. However, the share of cobalt was much higher in the emission profile of device 4 compared to all other systems. This leaves a question mark behind the result for cobalt.

Device 6 releases by far the highest PM mass (3 to 14 times more than the other devices). This results in the highest damage of all combustion systems with respect to PM_{2.5}. In exchange, the share of the UFP is smaller compared to the devices 1 and 4.

A general result for almost all combustion systems is that the ultrafine fraction accounts for the biggest part of the damage. Only for the devices 2 and 6 PM_{2.5} causes more damage

than the UFP`s. PM₁₀ is negligible for all combustion systems. This is a result of the clearly lower mass emitted as PM₁₀ and the lower CF of PM₁₀ compared to PM_{2.5}.

As in case study 2 there is – at least for some devices - considerable amount of the mass considered to be of unspecified chemistry. The mass shares of unspecified chemistry on the total PM mass vary between 0 and 94%. This leaves high uncertainties for those devices with high mass shares considered as unspecified chemistry (in particular device 5, less pronounced for device 1, 4 and 3).

There is a remarkable difference between the methods when comparing the performance of each device (Figure 28; log-scale). While EI99 and ReCiPe compute by far the highest damage for the device 6, the new method accounts for device 6 about the same damage potential as for device 5. On the other hand, the new method computes the highest damage for the device 4, while EI99 and ReCiPe calculate the 2nd best environmental performance for this device.

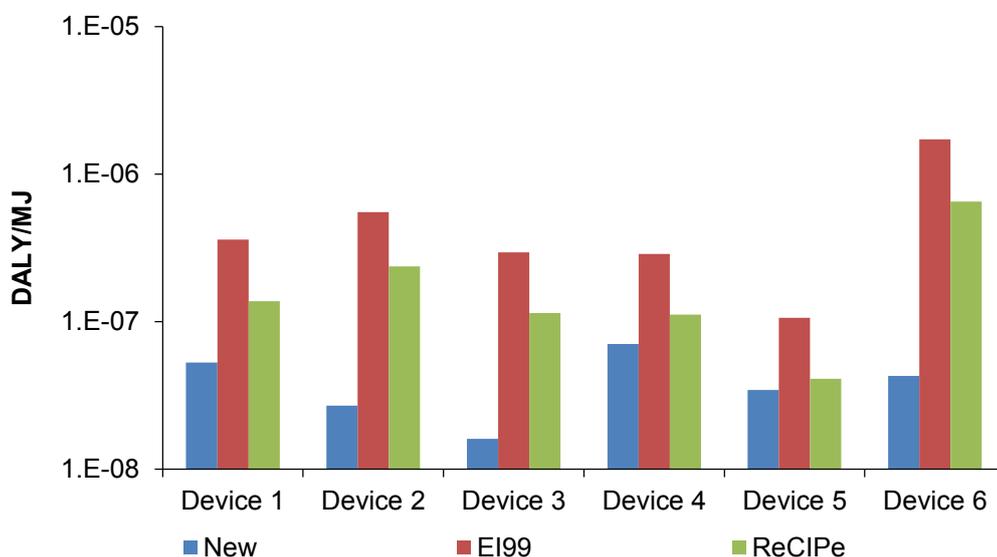


Figure 28. Comparison of the new impact assessment method with EI99 and ReCiPe.

This case study suggests the importance of the UFP in the impact assessment. Accounting for UFPs reveals to have major influence with respect to the overall ecological performance. The high number of UFPs (with low particle mass: ~10%) in device 4 accounts for the biggest damage to human health according to the new method.

Table 28 shows the total damage to human health due to PM assessed with EI99, ReCiPe and the new method and the damage ratio between the methods.

Damage in DALY/MJ	Device 1	Device 2	Device 3	Device 4	Device 5	Device 6
New	5.29E-08	2.69E-08	1.60E-08	7.06E-08	3.43E-08	4.28E-08
EI99	3.60E-07	5.52E-07	2.95E-07	2.87E-07	1.06E-07	1.72E-06
ReCiPe	1.37E-07	2.37E-07	1.15E-07	1.12E-07	4.10E-08	6.49E-07
% share						
New/EI99	15%	5%	5%	25%	32%	2%
New/ReCiPe	39%	11%	14%	63%	84%	7%

Table 28. Results in DALY/MJ heat produced 6 different devices for wood combustion assessed with 3 different IA methods and the share between different impact assessment methods.

All in all, in this study the new method computes smaller damage to human health than EI99 (2% - 32%) and ReCiPe (7% - 84%). Still, the difference in the results between the methods is not very big.

11.4 Case study 4: Mineral dust from sawing natural stone

Aim of this case study is to show effects of other kinds of particles originating from another source than wood combustion. For this reason a process was selected that emits a high amount of particles of a different chemical type. Sawing natural stone is a suited process for that. The process “1 kg natural stone plate, cut, at regional storage” is chosen from the ecoinvent database¹²⁹.

In order not to have a mixture of particles in the inventory, only particles from sawing process were considered. Particles from energy production, transport etc. were excluded in this study. The case study concentrates on the red box in the scheme below from Kellenberger¹²⁹.

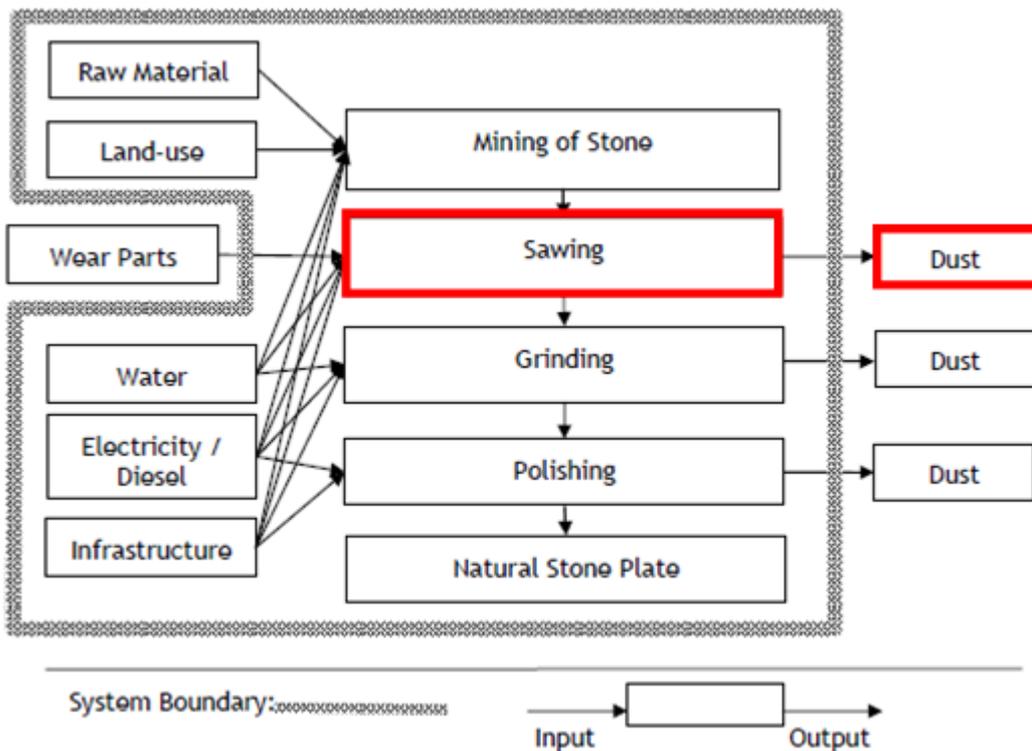


Figure 29. Flow diagram for the processing of the manufacture of natural stone plate taken from ecoinvent¹²⁹.

The total amount of particles in the air is assumed to be equal to the losses from cutting. The cuts are along the surface area, on the length and on the short edge. The slitting width is assumed to be 1 mm. The density of granite is 2750 kg/m³.

The share of the dust emission size categories is based on the EPA report “Generalized particle size distribution”¹³⁰ (<http://www.epa.gov/ttn/chief/ap42/appendix/appb-2.pdf>).

- 15% of the particulates are > PM₁₀
- 85% of the particulates are < PM₁₀
- 30 % of the particulates are < PM_{2.5}

95% of the particle mass is bound in the cooling water. The inventory contains the following values:

- PM_{10-2.5}: 0.001027 kg per kg granite
- PM_{2.5}: 0.00056 kg per kg granite

For these inventory data the CF for mineral dust were applied using the following assumptions: Solubility: insoluble, population density: low, stack height: ground level. The damage is expressed in DALY/kg cut natural stone (Table 29).

cutting natural stone			
Daly/kg	PM _{2.5}	PM ₁₀	Total
New	1.99E-13	7.12E-14	2.70E-13
EI99	3.97E-08	3.86E-08	7.84E-08
ReCiPe	7.84E-08	2.68E-08	4.13E-08
% share			
New/EI99	0.0005%	0.0002%	0.0003%
New/ReCiPe	0.0003%	0.0003%	0.0007%

Table 29. Results in DALY/kg cut natural stone and a comparison between different impact assessment methods.

Although the mass of PM is in the same range as in case study 3 the damage is about 5 orders of magnitude smaller. The reason for this is the very low effects for mineral dust on human health.

11.5 Case study 5: Heat from light fuel oil vs. heat from pellets

The goals of this case study are

- to provide a step by step procedure showing how the damage of PM in a process can be reassessed with the new method and implemented in an established life cycle impact assessment method (such as ReCiPe) by replacing only the damage to human health due to PM emission.
- to illustrate how double counting of damage to human health due to PM can be avoided as long as the new method is not incorporated in an established method.
- to find the potential significance of altered results when using the new impact assessment.

Within a complete comparative LCA, one subprocess (burning light fuel oil, burning wood pellets) which is assumed to have major influence on the LCA is modified in such a way that it can be used with the new method. All the other processes are not modified (transport, electricity production, infrastructure etc.; see Table 30).

For this purpose a case from the ecoinvent database was chosen that contains hazardous pollutants such as PAH, dioxins, heavy metals etc. These substances have to be deleted in the original assessment and then added back when assessed with the new impact assessment using the new CFs.

The product investigated is heat from pellets from the dataset “pellets, mixed, burned in furnace 15kW/MJ/CH”¹³¹ and it will be compared to the production of heat from light fuel oil “light fuel oil, burned in boiler 10kW condensing, non-modulating”¹³². The size of the heating systems is in a similar range. The light fuel oil boiler is a 10 kW boiler. The furnace for the pellets has 15 kW power. The ecoinvent datasets used for the foreground model for the production of heat from light fuel oil and pellets are listed in Table 30: The fuel burning process is highlighted in italic since only this dataset is modified for the new impact assessment. The functional unit is 1 MJ heat.

<u>Light fuel oil, burned in boiler 10kW</u>	<u>Pellets, mixed, burned in furnace 15kW</u>
<i>Light fuel oil, burned in boiler</i>	<i>Pellets, burned in furnace</i>
Light fuel oil, processing	Wood pellets, manufacturing
Electricity	Electricity
Oil boiler	Furnace, pellets (infrastructure)
Chimney	Transport
Oil storage 3000l	
Treatment, condensate	Disposal, wood ash mixture
Disposal, hazardous waste	Disposal, wood ash mixture

Table 30. Characterisation of the two types of heating.

The following assumptions have been taken for the case study:

- The PM is considered to be released in an urban location with a high population density, a low stack (typical for small combustion system of single family houses) and PM is of unspecified solubility.

Since there are no ready to use LCI for the new impact assessment method available, the inventory from ecoinvent had to be adapted in the following way:

- The total mass of PM_{2.5} from both combustion processes in ecoinvent is used as the starting point (PM₁₀ and UFP is not available in both ecoinvent combustion processes).
- The PNC from a pellet combustion process by Lamberg et al.¹²⁸ was used adjusting it as follows:

$$PNC(this\ study) = \frac{MassPM_{2.5}ecoinvent}{MassPM_{2.5}Lamberg} * PNC(Lamberg)$$

- In the same way, the amount of PM₁₀ was obtained:

$$Mass\ PM_{10}(this\ study) = \frac{MassPM_{2.5}ecoinvent}{MassPM_{2.5}Lamberg} * Mass\ PM_{10}(Lamberg)$$

- The process specific chemistry of PM from the ecoinvent datasets was assumed to be representative for all size classes (PM₁₀, PM_{2.5} and UFP).

Translating these assumptions into an inventory for the combustion processes results in a rather hypothetical emission profile (Table 31) with respect to the two chosen combustion processes fromecoinvent. However, the assumptions lead to an inventory which contains all size classes and almost 20 different chemical species and therefore reflects a case which could depict a realistic inventory specifically elaborated for the new impact assessment.

	Light fuel oil, burned in boiler 10kW			Pellets, mixed, burned in furnace 15kW		
	PM ₁₀ (kg/MJ)	PM _{2.5} (kg/MJ)	UFP (#particles/MJ)	PM ₁₀ (kg/MJ)	PM _{2.5} (kg/MJ)	UFP (#particles/MJ)
Carbon black				8.6E-08	4.1E-07	1.6E+12
Ammonia				3.6E-07	1.7E-06	6.6E+12
Sulfur dioxide	7.9E-06	3.8E-05	1.7E+14	5.2E-07	2.5E-06	9.5E+12
Arsenic				3.9E-11	1.9E-10	7.1E+08
Cadmium				1.5E-10	7.0E-10	2.7E+09
Chromium VI				3.5E-10	1.7E-09	6.3E+09
Copper	8.3E-11	4.0E-10	1.5E+09	5.6E-09	2.7E-08	1.0E+11
Lead				3.4E-09	1.6E-08	6.2E+10
Manganese				1.7E-08	8.3E-08	3.2E+11
Mercury	1.0E-10	5.0E-10	1.9E+09	6.3E-11	3.0E-10	1.1E+09
Nickel				8.6E-11	4.1E-10	1.6E+09
Cobalt				8.6E-11	4.1E-10	1.6E+09
Zinc	1.0E-10	5.0E-10	1.9E+09	7.2E-08	3.5E-07	1.3E+12
Vanadium				8.6E-11	4.1E-10	1.6E+09
Iron				1.2E-09	5.8E-09	2.2E+10
monocyclic aromatic hydrocarbons	4.2E-09	2.0E-08	7.6E+10			
Alkane/Alkene/Alkine	5.6E-08	2.7E-07	1.0E+12	8.4E-07	4.0E-06	1.5E+13
polycyclic aromatic hydrocarbons	9.6E-11	4.6E-10	1.8E+09	1.5E-10	7.1E-10	2.7E+09
Dioxin	1.2E-17	5.7E-17	2.2E+02	6.5E-15	3.1E-14	1.2E+05
unspecific	4.3E-08	2.1E-07	7.9E+11	1.6E-05	7.7E-05	2.9E+14
Total	8.0E-06	3.8E-05	1.8E+14	1.8E-05	8.6E-05	3.3E+14

Table 31. Inventory of the emissions for both combustion processes.

In the following, a step by step procedure is provided showing how the damage of PM from the heat production (light fuel oil or pellets) can be reassessed with the new method and implemented in existing life cycle impact assessment methods such as ReCiPe:

- Into the processes building the foreground model (Figure 30), then
- Into the fuel burning process. The damage is split in midpoint indicators (Figure 31).
- Next, the damage due to PM is assessed with the new IA method for the pellet burning process (Figures 32 and 33) and the light fuel oil combustion process (Figure 34 and 35).
- The damage of PM assessed with ReCiPe has to be replaced with the damage assessed with the new method (Figure 36).
- The net damage is added back to the fuel burning process (Figure 37).
- Finally, the adopted fuel burning process is added back to the life cycle damage (Figure 38).

Figure 30 shows the total damage in points per MJ heat produced for both processes using the impact assessment method ReCiPe. The damage is split into the processes building the foreground model. Interestingly, the production of light fuel oil causes more damage than all the other processes together, including burning light fuel oil. The same is true for the system with pellets.

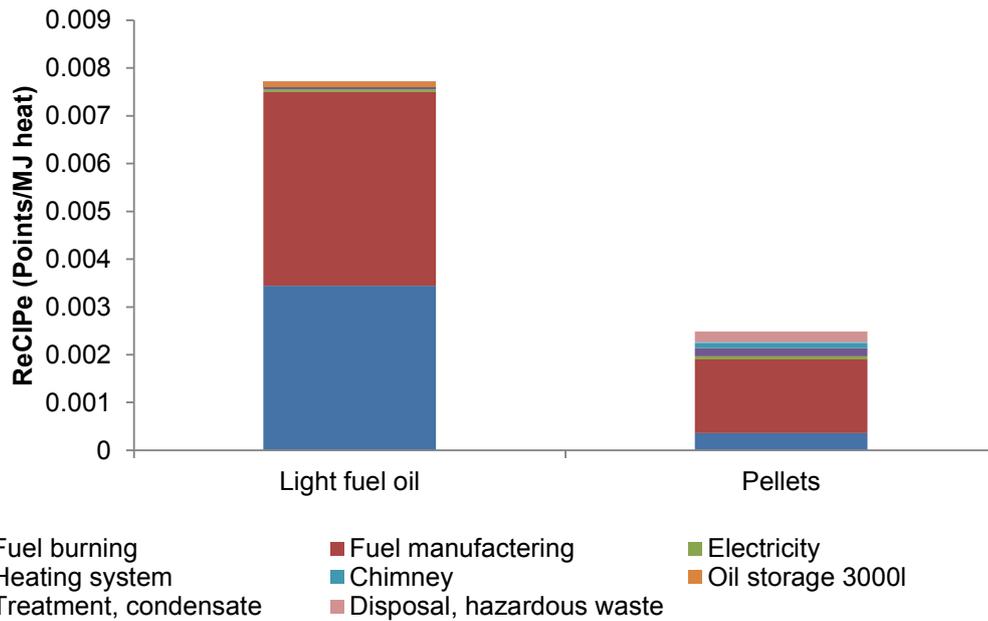


Figure 30. Total contribution of all processes assessed with ReCiPe.

In a next step, it is focussed on the fuel burning process (blue bar, 0.0035 points for light fuel oil, 0.0003 points for pellets). In order to detect the damage due to PM for the fuel burning process the contribution of the different impact categories has to be assessed. The damage due to particles is accounted for in the category “Particulate matter formation” (Figure 31).

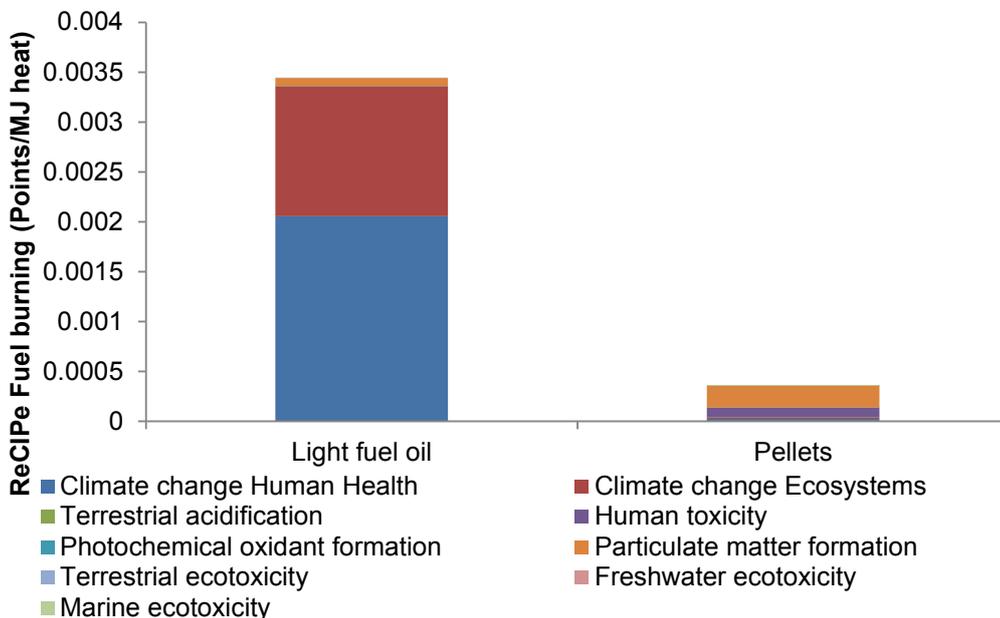


Figure 31. Total damage depicted with respect to all the impact categories, but for the fuel burning process only.

Climate change is the major contributor to the total damage in the light fuel oil process. Particulate matter formation and Human toxicity have significant shares in the pellets process, but only very small share on the light fuel oil process. The damage due to PM is bigger (orange part of the bar) in the Pellets process compared to the Light fuel oil process.

The Particulate matter formation and Human toxicity impact categories will be reassessed with the new impact assessment model. Therefore, the life cycle inventory model for pellets and oil burning process are adapted. Table A6 and A7 in the Annex indicates all substances that are assumed to be particulates labelling them with “P”. These substances were deleted in the original inventory and are assessed with the new method. By deleting these substances in the original inventory double counting of particulate emissions can be avoided.

Since the inventory in ecoinvent does not contain number concentration of UFP, data from the pellets burning process in case study 3 were adapted. The UFP number concentration was linear adapted to the amount of PM_{2.5} mass. It is assumed that all size fractions have the same chemical profile.

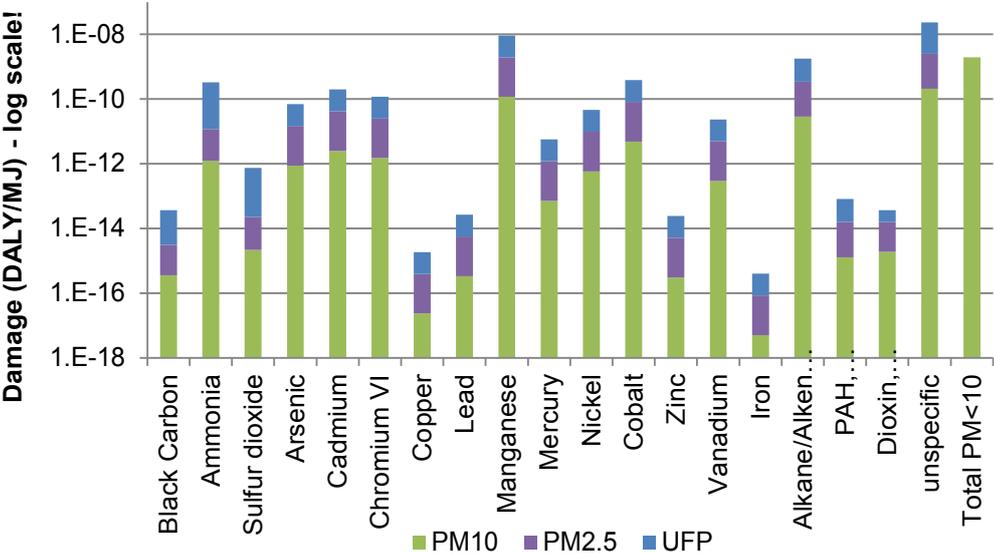


Figure 32. Size segregated damage on a log scale for the pellet burning process.

Figure 32 shows chemically disclosed and size segregated results for the pellets process. Figure 32 and Figure 33 show the same data. Figure 32 depicts the contribution of all size fractions using a log scale for the damage axis, while Figure 33 highlights the dominant contribution of the UFP fraction using a normal axis. This result could be expected since the share of PM_{2.5} and UFP is the same as in case study 3 where we found the UFP to be dominant. As found in case study 2 and partly in case study 3, the unspecified fraction plays an important part. It seems that chemical disclosure of the particle mass is a possible bottleneck for the outcome of the results using the new impact assessment.

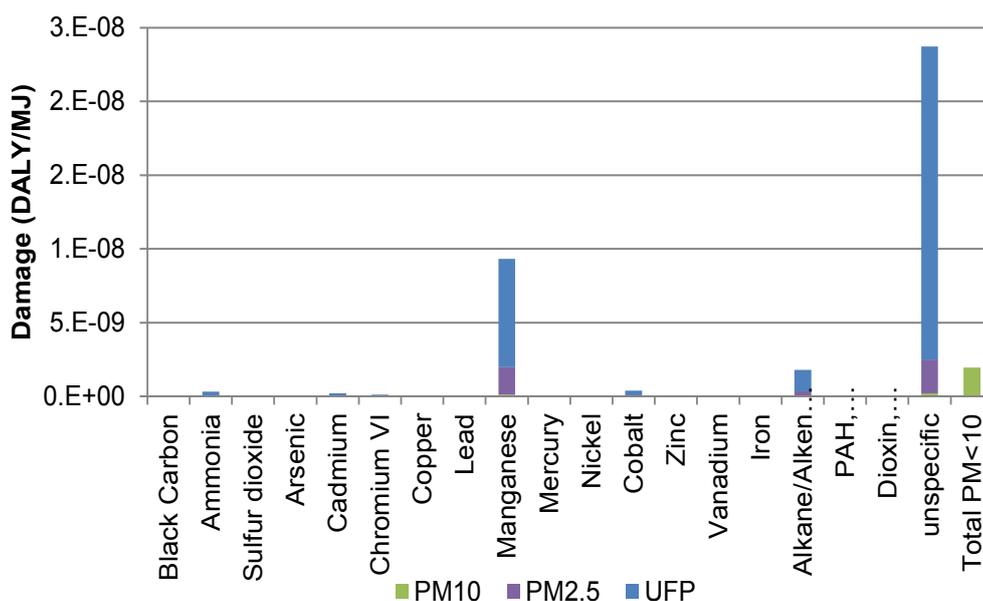


Figure 33. Size segregated damage on a normal scale for the pellet burning process.

Beside the unspecified fraction, manganese is the 2nd substance that causes high damage in the pellets combustion process. It might be a surprise to find manganese having a higher damage potential than PAH or dioxin. This might be explained by the high quality of the burning conditions in modern pellet furnace.

Table 32 lists detailed impact assessment results presented in the Figures 33 and 34 in order to decipher the contribution of all substances, including those which contribute so little that their share is not visible in the Figure.

Damage (DALY)	PM ₁₀	PM _{2.5}	UFP	Total
Black Carbon	3.61E-16	2.79E-15	3.32E-14	3.63E-14
Ammonia	1.22E-12	1.06E-11	3.19E-10	3.31E-10
Sulfur dioxide	2.18E-15	2.06E-14	7.31E-13	7.53E-13
Arsenic	8.78E-13	1.40E-11	5.51E-11	6.99E-11
Cadmium	2.49E-12	3.96E-11	1.56E-10	1.98E-10
Chromium VI	1.50E-12	2.38E-11	9.40E-11	1.19E-10
Copper	2.34E-17	3.73E-16	1.47E-15	1.87E-15
Lead	3.32E-16	5.28E-15	2.08E-14	2.64E-14
Manganese	1.17E-10	1.86E-09	7.34E-09	9.31E-09
Mercury	7.06E-14	1.12E-12	4.43E-12	5.63E-12
Nickel	5.84E-13	9.30E-12	3.67E-11	4.66E-11
Cobalt	4.88E-12	7.76E-11	3.06E-10	3.89E-10
Zinc	3.03E-16	4.82E-15	1.90E-14	2.41E-14
Vanadium	2.92E-13	4.65E-12	1.83E-11	2.33E-11
Iron	5.05E-18	8.04E-17	3.17E-16	4.02E-16
Alkane/Alkene/Alkine	2.85E-11	3.20E-10	1.45E-09	1.80E-09
PAH	1.30E-15	1.46E-14	6.60E-14	8.19E-14
Dioxin	1.90E-15	1.40E-14	2.06E-14	3.64E-14
unspecific	2.10E-10	2.28E-09	2.12E-08	2.37E-08
Total	3.67E-10	4.64E-09	3.10E-08	3.60E-08

Table 32. Impact assessment results for the pellet burning process.

The same analysis is performed for the light fuel oil combustion process. Figure 34 and 35 depict chemically disclosed and size segregated results for the light fuel process. The total damage is much smaller compared to the pellets process. Obviously there are less substances building up particle mass. This indicates either that the quality of the LCI is not as good as for the pellets process, or the light fuel oil at this stage contains much less chemical substances than wood pellets.

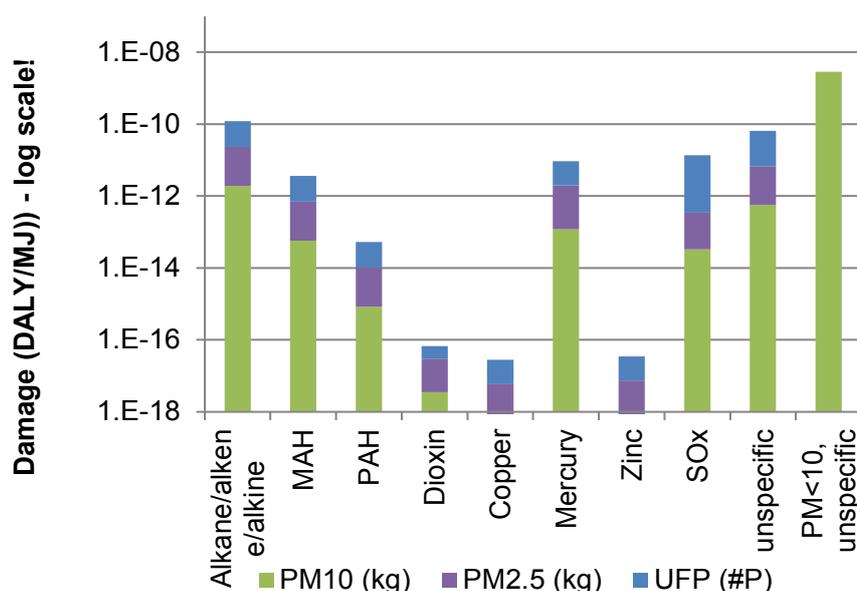


Figure 34. Size segregated damage on a log scale for light fuel oil burning process.

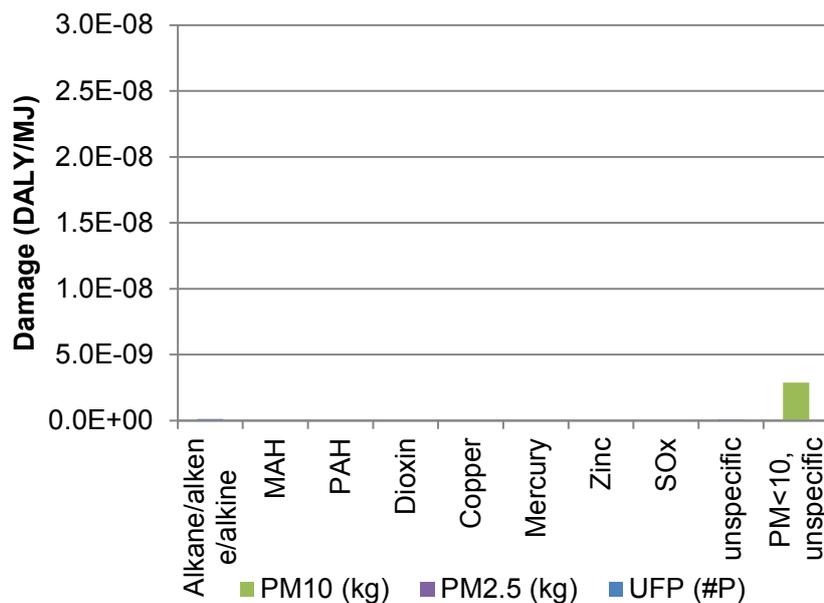


Figure 35. Size segregated damage on a normal scale for the light fuel oil burning process.

Table 33 lists detailed impact assessment results presented in the Figures 35 and 36.

Damage (DALY)	PM ₁₀	PM _{2.5}	UFP	Total
Alkane/alkene/alkine	1.92E-12	2.16E-11	9.74E-11	1.21E-10
MAH	5.70E-14	6.42E-13	2.90E-12	3.60E-12
PAH	8.37E-16	9.42E-15	4.26E-14	5.28E-14
Dioxin	3.49E-18	2.57E-17	3.78E-17	6.70E-17
Copper	3.49E-19	5.55E-18	2.19E-17	2.78E-17
Mercury	1.18E-13	1.87E-12	7.39E-12	9.38E-12
Zinc	4.36E-19	6.94E-18	2.74E-17	3.47E-17
SOx	3.29E-14	3.12E-13	1.33E-11	1.37E-11
unspecific	5.68E-13	6.17E-12	5.75E-11	6.42E-11
Total	2.69E-12	3.06E-11	1.79E-10	2.12E-10

Table 33. Impact assessment results for the light fuel oil burning process

The next step to do in order to replace the impact assessed with ReCiPe with the impact assessed by the new method is deleting the substances declared as being particular in the category of human toxicity and adding back the impact for the same substances using the new impact assessment method. Of course, this is done for both processes. Figure 36 depicts the results step by step.

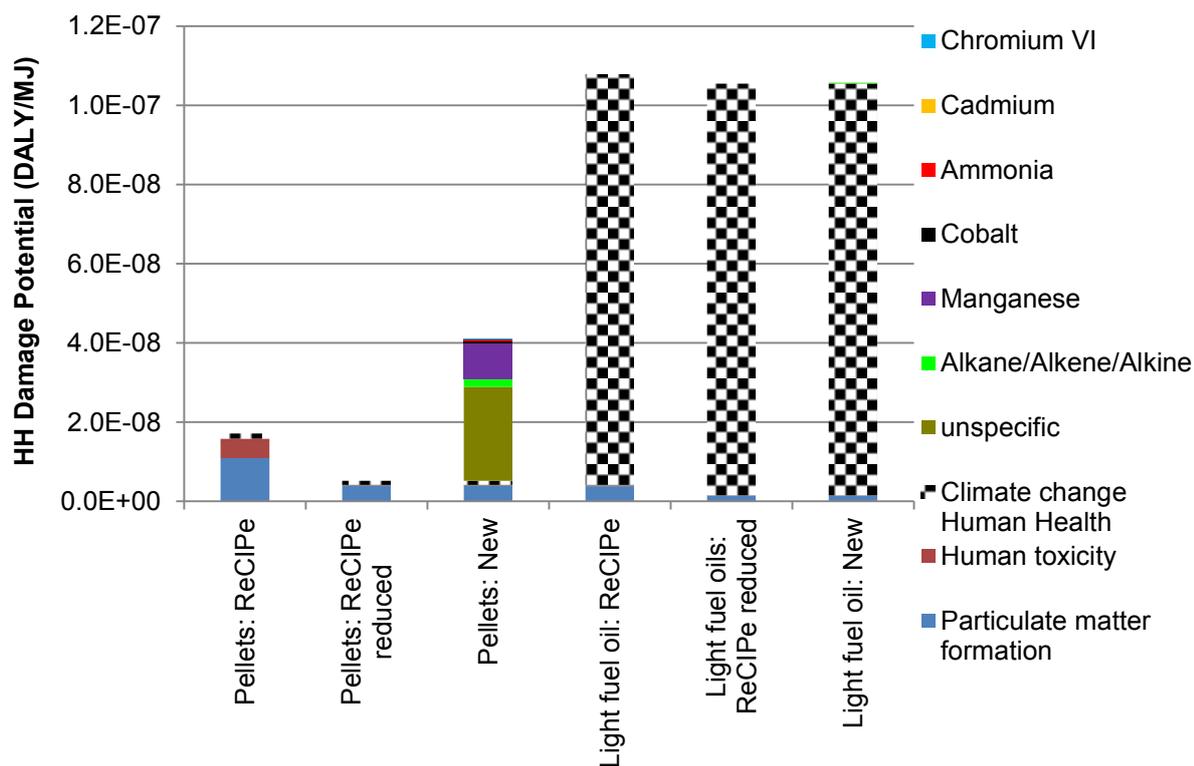


Figure 36. Step by step: Original impact of Particles – reduced impact after deleting particulate substance – adding back for both processes.

The first colon depicts the original damage to human health (endpoint in DALY/MJ) calculated with ReCiPe for the fuel burning processes. Most impact assessment categories are not visible, since they are not relevant for the burning process (e.g. marine toxicity, ozone depletion, etc.). The 2nd colon depicts residual damage after having deleted all substances considered to be particulates (all substances labelled with P in the life cycle inventory). The 3rd colon depicts the damage after adding back the damage of the particulate substances.

With respect to the pellets fuel burning the new impact assessment computes more than twice the original impact. In the light fuel oil process the total damage is slightly smaller after reassessment with the new method compared to the original assessment with ReCiPe. Anyway, the total damage of the light fuel oil is dominated by impacts due to climate change.

One more step backwards (Figure 37) reveals the overall damage throughout all impact categories within ReCiPe. The damage is now expressed in points (as in Figure 30 and 31) since several endpoint categories are summarized in one single score indicator.

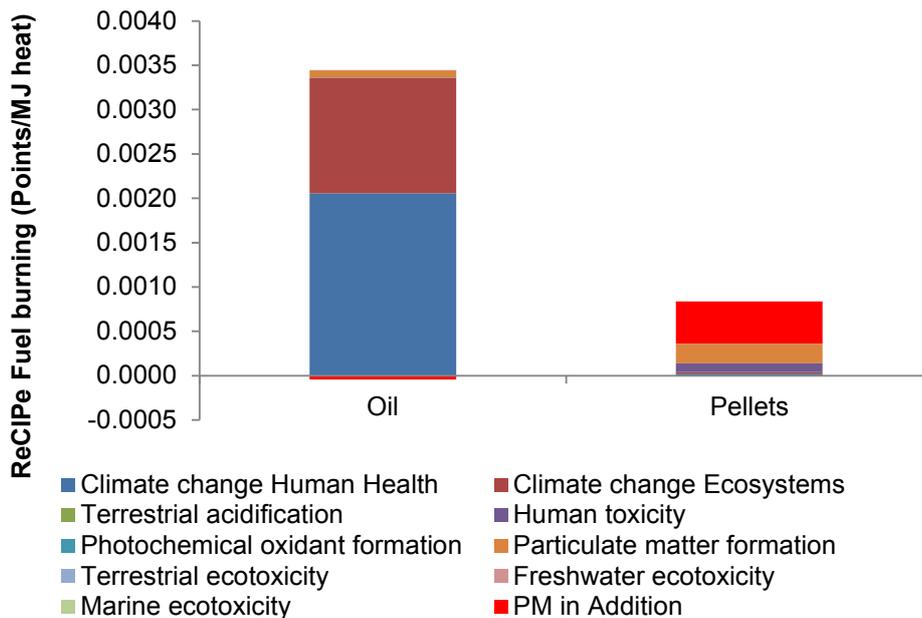


Figure 37. Total damage depicted with respect to all the impact categories, but for the fuel burning process only.

The total damage of the light fuel oil burning process decreases slightly. This is indicated with the small, red share below 0. With regard to the overall performance for Pellets burning process the results from the new impact model lead to damage twice as high after the reassessment.

Finally, the results for the new assessment on the level of the processes are shown in Figure 38.

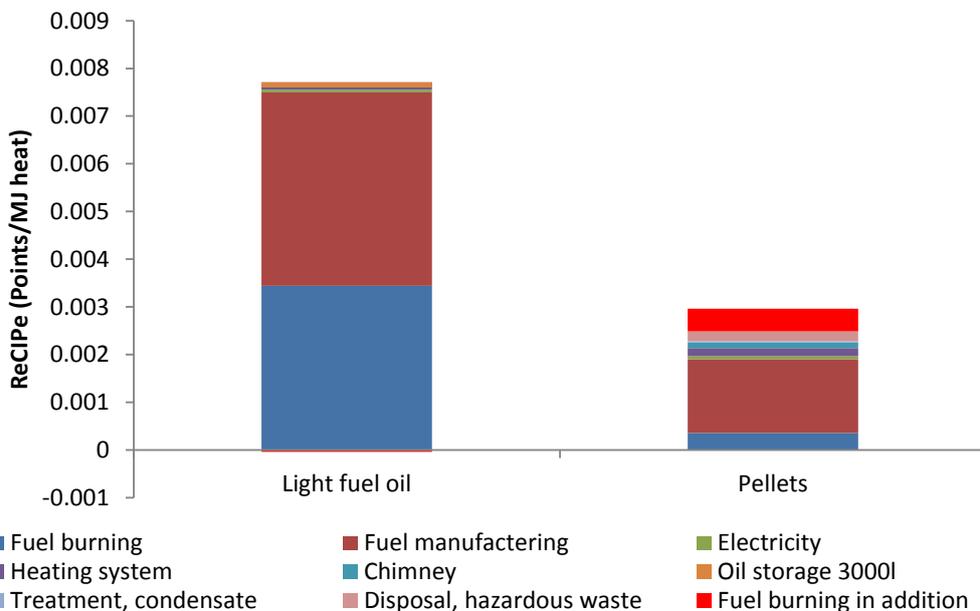


Figure 38. Total contribution of all processes after reassessment.

As expected the same (although diminished) trend was also found on the level of processes: a very small reduction of the impacts for the light fuel process and an increase of the total impacts for the pellets process.

Table 34 shows the total damage to human health due to PM assessed with ReCiPe and the new method and the damage ratio between the methods.

Damage in DALY/MJ	Pellets	Oil
New	3.60E-08	2.13E-10
ReCiPe	1.59E-08	4.00E-09
% share		
New/ReCiPe	226%	5%

Table 34. Results in DALY/MJ heat produced by burning pellets or fuel and comparison between different impact assessment methods for PM.

This case study shows that splitting of PM in size fractions and chemical compositions and replacing the damage in a complete LCA may lead to a net benefit (for the light fuel oil process) or a drawback (for the pellet process).

To summarise, the damage to human health increases for the wood pellets burning process while the damage decreases for the light fuel oil burning process. This result should not be overstated neither should it be generalized for future application. The life cycle inventories modelled might be too rough for such a conclusion (the share of chemically unspecified substances is rather big and the derivation of the three size fractions rely on other processes).

The case study provides a step by step procedure in order to replace damage to human health due to PM and is therefore an important proof of concept for the future applicability of the new method.

The case study further illustrates exemplarily how double counting of metals, PAH, dioxins etc. can be avoided.

11.6 The new impact assessment versus the established ReCiPe and Ecoindicator 99 methods

Considering the base case setup applied for all case studies (with respect to population density, stack height and solubility) it is possible to compare the results of the new impact assessment method with the results from established methods such as ReCiPe and Ecoindicator 99 (Table 35).

case 1: comparing different types of wood						
PM _{2.5} only	Typ 1	Typ 2	Typ 3	Typ 4	Typ 5	
New/EI99	32%	100%	95%	83%	69%	
New/ReCiPe	88%	274%	259%	228%	188%	

case 2: comparing an old and a new stove		
Pm ₁₀ and PM _{2.5}	stove 1	stove 2
New/EI99	4%	3%
New/ReCiPe	11%	8%

case 3: small scale wood combustion						
PM ₁₀ , PM _{2.5} and UFP	Device 1	Device 2	Device 3	Device 4	Device 5	Device 6
New/EI99	15%	5%	5%	25%	32%	2%
New/ReCiPe	39%	11%	14%	63%	84%	7%

case 4: sawing natural stone plate	
PM ₁₀ and PM _{2.5}	
New/EI99	0.0003%
New/ReCiPe	0.0007%

case 5: heat from Oil vs. wood pellets		
PM ₁₀ , PM _{2.5} and UFP	Pellets	Oil
New/ReCiPe	226%	5%

Table 35. Comparison of established and the new impact assessment in an overview over all case studies.

Wood burning systems lead to similar results with the established EI99 and ReCiPe and the new impact assessment. The values in Table 35 are calculated as the damage calculated with the new method (New) divided by the damage calculated with an established method (EI99, ReCiPe). For all cases of wood combustion, “New” calculates on average less damage than EI99 (values always < 100%). For ReCiPe the values vary between 7% and 274%.

With regard to case study 4 (accounting or mineral dust) New assess damage about 6 orders of magnitude smaller than EI99 and ReCiPe.

12 Monetarisisation of damage – valuation of a life year (VOLY)

In economics, an externality is the cost or benefit that affects a party who did not choose to incur that cost or benefit. A negative externality (also called "external cost" or "external diseconomy") is an action of a product or services on consumers that imposes a negative effect on a third party; it is "external cost". Many negative externalities are related to the environmental consequences of production and consumption of goods and services. For example, manufacturing activities that cause air pollution impose health and clean-up costs on the whole society. Air pollution from burning fossil fuels causes damages to crops, (historic) buildings and public health, etc. The most extensive and integrated effort to quantify and monetise this impact was in the European ExternE project series.

The ExternE project developed a framework in order to transform impacts that are expressed in different units into a general unit (monetary value). In order to calculate the external costs of polluting activities one needs to carry out an impact pathway analysis. Figure 39 is taken from the ExternE report¹³³ and depicts the main steps of an impact pathway analysis.

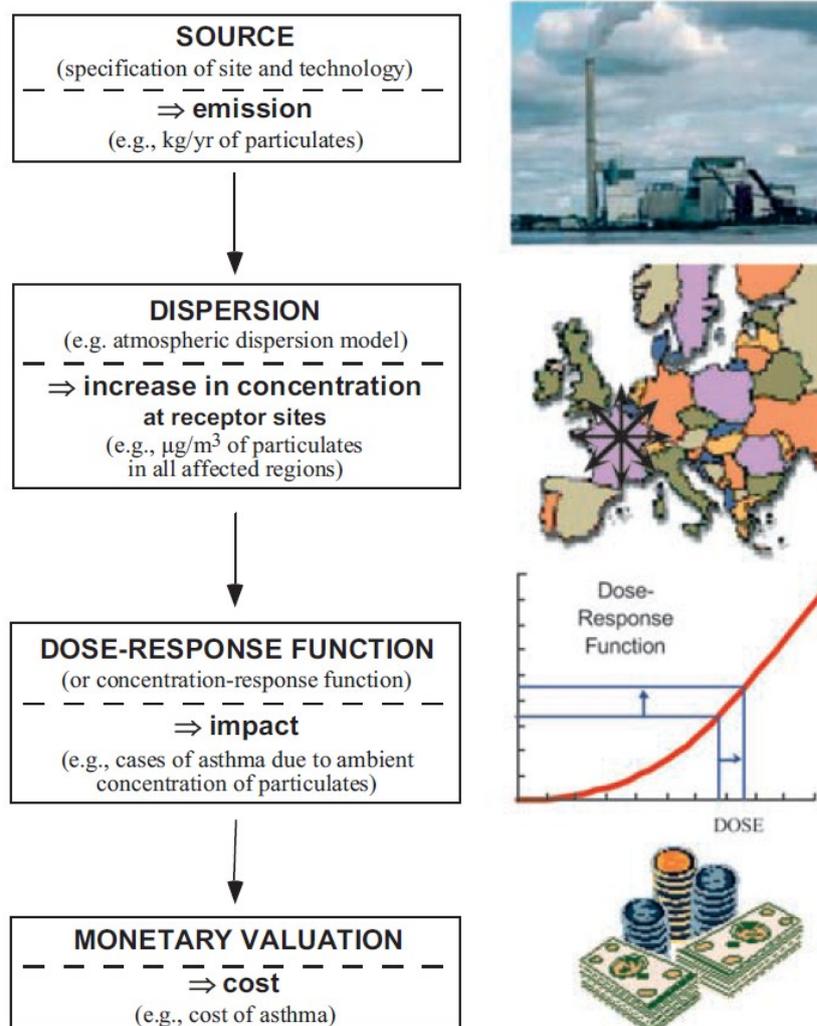


Figure 39. The figure taken from ExternE depicts the impact pathway analysis¹³³.

The single steps of the impact analysis pathway can be grouped as follows:

- Emission: specification of the relevant technologies and pollutants, e.g. kg of PM per reference flow (e.g. 1 kWh) emitted by a power plant.
- Dispersion: calculation of increased pollutant concentrations in all affected regions (this step is also called environmental fate analysis).
- Impact: calculation of the dose from the increased concentration, followed by calculation of impacts (damage in physical units) from this dose, using a dose-response function, e.g. cases of lung cancer due to this increase in PM.
- Cost: economic valuation of these impacts, e.g. multiplication by the cost of a case of lung cancer.

The method applied in ExternE resembles very much the method for life cycle impact assessment. This allows combining the new impact assessment with the impact pathway analysis from ExternE. The steps emission specification, dispersion (fate) exposure and impacts (combining exposure, effects and damage) from the new impact assessment can be used and directly combined with the valuation of the economic costs used in ExternE.

The value of life years (VOLY) is assessed considering mortality and morbidity. As the starting point for the valuation of health end-points is the identification of the components that comprise changes in welfare. These components should be summed to give the total welfare change, assuming no overlap between categories. The three components include:

- Resource costs i.e. medical costs paid by the health service in a given country or covered by insurance, and any other personal out-of-pocket expenses made by the individual (or family).
- Opportunity costs i.e. the cost in terms of lost productivity (work time loss (or performing at less than full capacity)) and the opportunity cost of leisure (leisure time loss) including non-paid work.
- Disutility i.e. other social and economic costs including any restrictions on or reduced enjoyment of desired leisure activities, discomfort or inconvenience (pain or suffering), anxiety about the future, and concern and inconvenience to family members and others.

The cost items covered include:

- Health care resource costs
- Costs of absenteeism
- Hospital admissions
- Emergency-room visits for respiratory illness
- Visit to a doctor: asthma and lower respiratory symptoms
- Restricted activity days
- Respiratory symptoms in adults and children with asthma
- Respiratory medication use by children and adults
- Chronic bronchitis

The value of a VOLY for European conditions is assumed to be 41'000 €^{134, 135}. Since the VOLY and the DALY are derived in a similar way, it can be assumed that the VOLY is a suitable measure for the value of a DALY¹³⁴.

The damage in DALY calculated with the new impact assessment, ReCiPe and Ecoindicator 99 can now be translated in economic damages using the 41'000 €/DALY. Table 36 shows for all the case studies the damage to human health and the economic damage as a function of the respective functional unit.

case study 1						
Damage in €/kWh	Typ 1	Typ 2	Typ 3	Typ 4	Typ 5	
EI99	0.008	0.023	0.010	0.013	0.007	
ReCiPe	0.003	0.008	0.004	0.005	0.003	
New	0.002	0.023	0.009	0.011	0.005	

case study 2		
Damage in €/kg wood combusted	stove 1	stove 2
EI99	0.217	1.198
ReCiPe	0.080	0.439
Total	0.008	0.049

case study 3						
Damage in €/MJ	Device 1	Device 2	Device 3	Device 4	Device 5	Device 6
New	0.002	0.001	0.001	0.003	0.001	0.002
EI99	0.015	0.023	0.012	0.012	0.004	0.071
ReCiPe	0.006	0.010	0.005	0.005	0.002	0.027

case study 4	
Damage in €/kg cut stone	
New	0.00000001
EI99	0.003
ReCiPe	0.002

case study 5		
Damage in €/MJ	Pellets	Oil
New	0.001	0.00001
ReCiPe	0.001	0.00016

Table 36. Economic damage of PM for all case studies as a function of the respective functional unit.

The interpretation of this result is somewhat difficult. For that reason in the following an illustrative example is given. According to the Swiss Federal Statistical Office the final energy consumption in Switzerland produced from wood or charcoal sums up to 22'416 TJ¹³⁶. Case study 3 shall be used in order to get a rough estimate on the economic consequence using established or new IA methods, assuming that all the heat energy is produced with two devices selected from case study 3. Case study 3 reveals that device 5 assess the smallest difference between the new and the established IA methods. Contrariwise, device 6 assesses the biggest difference between the new and the established IA methods. Table 37 summarises the range of possible economic impacts under the conditions mentioned.

Deviation in € for production of 22'416 TJ heat	minimum	maximum
between New and EI99	6.6.E+07	1.5.E+09
between New and ReCiPe	6.1.E+06	5.6.E+08

Table 37. Economic differences between assessments using the new and established methods.

In the best case the difference of the economic impacts could be around 6 million Euros, the worst case would be 1.5 billion Euros. The example illustrates how economic impacts could be assessed. However, the result shall not be interpreted as a realistic scenario since there are crucial limitations included, for example:

- it relies on artificial LCI with rather high uncertainties as discussed in case study 3.
- it is assumed that all the energy produced with wood and charcoal is produced with one of these two stoves.
- it is assumed that all the emissions take place in urban area with high population density.

13 Perspective: New impact assessment and the ecological scarcity method

The ISO 14044 standard defines Life Cycle Impact Assessment (LCIA) as the “phase of life cycle assessment aimed at understanding and evaluating the magnitude and significance of the potential environmental impacts of a product system”¹³⁷. The aim of the impact assessment phase is to translate the life cycle emissions (to air, water and soil) and resource consumption (material and energy) from the life cycle inventory into environmental impacts for the Areas of Protection, i.e. to assess the impact on the entities that we want to protect. The Areas of Protection considered in the ILCD Handbook are Human Health, Natural Environment and Natural Resources¹⁰. The ILCD Handbook further states that “for impacts on Human Health caused by various types of environmental stressors, the aim is to quantify the changes in both mortality and morbidity that are associated with goods or services in an integrated way.” And as the most appropriate metric to integrate the various stressors for the Area of Protection Human health the DALY concept is recommended. Modelling the environmental mechanism (cause effect chains) of PM requires the framework for a fate, exposure and effect analysis.

The present work followed these recommendations of the ILCD and adopted the DALY as the measure to interpret the damage to human health due to PM. Contrariwise the ecological scarcity method takes another perspective. In the ecological scarcity method, an impact assessment of life cycle inventories is performed according to the “distance to target” principle. This means that the difference (distance) between the current flow (current annual pollutant load or resource extraction) is set in relation to the critical flow (critical annual pollutant load or resource extraction) in a specific area. The result is expressed in eco-points per unit of pollutant emission or resource extraction. Thus, the ecological scarcity method does not model a cause effect chain and therefore is not split in fate, exposure and effect analysis.

The two impact assessment method follow different modelling principles and the units of the results are expressed in not comparable units. This, together with the fact that political targets for different PM size fractions are missing lead to the conclusion that the results from the new impact assessment model (including intermediate results for fate, exposure, effects of size, solubility and chemistry) cannot be included in the ecological scarcity method.

However, it may be possible to adapt the ecological scarcity method based on its own modelling principles on comparing current flows with critical flows as it is done already for a variety of substances (e.g. PAH, dioxin) that are components of PM. Three major problems may arise in case of an adaptation of the ecological scarcity method to the requirements of the new impact assessment.

- The legislation in the specific geographic area may probably not provide critical flows for PM emissions segregated into size fractions. The ecological scarcity method depends on the availability of critical flows. In case there are no critical flows available for some types of particles the ecological scarcity method could adapt a simplified system. For example the Swiss law nowadays provides critical

flows only for PM₁₀, but not for PM_{2.5} and UFP. The ecological scarcity method could then just neglect the fact that PM is segregated into sizes and just assess PM₁₀ potentially separated into chemicals. Similar to the effect of size the effects of solubility may also not be neglected in case of missing critical flows considering solubility. Neglecting the effects of different sizes or solubility is a simplification of the new impact assessment.

- Many processes release PM emissions that are not very toxic, for example processing agricultural land, PM from cutting of refining natural stone, etc. It is very likely that regulatory authorities do not provide critical flows for many emissions. The ecological scarcity method could either neglect this type of emissions or assess the emission using unspecified chemistry (basically as it does today). Both ways reflect a simplification. Neglecting this type of emissions may lead to unfair results in the case of a comparative LCA in which one process by chance releases PM where the legal authority provides critical flow and the comparative process releases PM but the legal authority does not provide critical flows. Instead of neglecting the emission, applying the ecological scarcity method one could try to assess as much of the PM emissions as possible and assess the rest of the mass as an unspecified chemistry of PM. However, following such a strategy may overestimate the total impact of PM since one could assume that for the most hazardous substances the legal authority is able to provide critical flows, but for the less hazardous part of PM there are probably no critical flows available. In fact, a relevant mass share of PM is of low toxicity (e.g. sea salt, mineral dust). Allocating the mass of these substances to the CF for PM₁₀ with unspecific chemistry would overestimate the total impact of PM₁₀.
- Among many organic substances dioxins and PAH or some metals (lead, cadmium, zinc) are separately assessed in the ecological scarcity method. Under ambient conditions these substances are always in a solid state and thus contribute to the PM mass. If the critical flows for PM provided by regulatory authority are based on epidemiological studies to calculate the critical flow, there will be double counting for substances as those mentioned above.

Summarized, a simplified adaptation of ecological scarcity provides on one hand the benefits of a more detailed analysis of PM emissions into the air (better representation of damage to human health, improved reporting due to the resolution of the results in sizes classes, chemical substances etc.). The importance of a revision of impact assessment methods for PM is discussed in the introduction. On the other hand, partly incorporating this system may lead to other problems with respect to the application of the method (fair comparison of the impacts and reporting this transparently). Neglecting those chemical substances for which no critical flows are available may lead to unfair comparison and assessing emissions for unspecified chemistry due to a lack of critical flow parameters may overestimate the real impact of such an emission.

An alternative strategy for the ecological scarcity may be not to adopt the ideas of the new impact assessment. This may be justified by reasoning that the ecological scarcity benchmarks current emission flows on critical flows set by a legal authority. In case the legal authority provides only critical flows for, e.g. PM₁₀, as in Switzerland, there is no

demand for a more detailed assessment of PM₁₀. Hence, it is important that the Ordinance on Air Pollution Control (OAPC) contains more objectives for specific sizes of PM.

As long as life cycle inventories are not adopted in a way suitable for the new impact assessment the ecological scarcity method can be used unaltered and without any limitations. Even if life cycle inventories will be adapted in the future suitable for the new impact assessment method (PM emissions split in size categories, chemical substances, etc.) the ecological scarcity method may continue to work as it does today. The only limitation compared to today will be that in a life cycle inventory all the mass that contributes to the total PM emissions has to be summed up. PM can then be assessed as PM₁₀ as it is done today.

14 Project resume and outlook

This project successfully built up a new impact assessment model based on chemical and physical properties of PM by substantially improving fate, exposure and, in particular, the effect model. However there are some limitations in the model and with respect of using applying the model. Therefore it is suggested to initiate a few follow up projects in the near to medium term.

14.1 Achievement of objectives

Finally, the new impact assessment accounts for:

- Size: with 3 levels (PM₁₀, PM_{2.5}, UFP)
- Amount of PM: measuring mass (kg) for PM₁₀ and PM_{2.5}, and Measuring number concentration (#Particles) for UFP
- Solubility: with 3 levels (soluble, not soluble, unspecified solubility)
- Chemistry: with 34 different chemicals
- Population densities: with 3 different levels (city centre, high and low population density)
- Site specification: with 3 types of sites (kerbside, urban, rural)
- Stack height: with 3 levels of height for the emission release (ground level, low and high stack)

This leads to total amount of 2781 CF. The high number of CF allows accounting for a very high accuracy (if the inventory is of good quality) and very high resolution (visibility for the evaluation) what facilitates the interpretation of future LCA results with special focus on PM emissions.

There are numerous additional benefits from the project to be mentioned:

- The literature available for fate of PM in the atmosphere with respect to LCA modelling and literature of epidemiological studies with respect to different size effects and chemical composition has been summarised and structured.
- Beside chemical and physical properties (size, amount, chemistry, solubility) the new impact assessment accounts for further relevant information, such as site specific information for the release of the emission, the height above ground for the release of the emissions or population density in the area of the release of the emission.
- Based on the literature studies for fate, exposure and effects on human health, the models were adapted and significantly improved. For example, the new fate model is to our knowledge the first one accounting for coagulation processes for UFP in LCA application and, in addition, models fate and exposure depending on particles sizes from 1 nm to 10 µm.
- Systematic double counting (of particulate substances) occurring in established method can be eliminated as shown in case study 5.
- All models are dynamically linked. Thus, the models can be adapted to new requirements (e.g. new setting of parameters for a new geographical region,

developing new CF for additional substances, taking account of new chemical and or physical properties, engineered nanoparticles, etc.).

- The entire model is based on Microsoft excel. This allows adaptation of the tool at a later date without requiring special knowledge.
- The functionality of the model is verified and allows for differentiated assessments as long as the LCI data represent the requirements of the tool. First results from the case studies show the expected differences between the new and established methods comparing different PM emissions.

However, there are some limitations when using the new impact assessment method.

14.2 Limitations of the new impact assessment

Despite strong adaptations of established impact assessment models there are still some shortcomings and limitations:

- There is still a considerable amount of uncertainty reflected in the toxicity of the chemical substances. This can be explained because the data on toxicity stem from toxicological studies. Ideally, one would use only epidemiological studies to calculate the effect on humans due to the chemistry of various PM components. Such data, however, will probably not be available in sufficient quantity in the next 10 - 20 years.
- One important category of fine dust - the secondary organic PM - could not be implemented. In collaboration with experts from atmospheric chemistry, atmospheric physics and climate modelling, we had to conclude that the state of research is not progressed enough to implement secondary organic particles in our model today.
- There are a number of processes which are not considered in this model. For example, nucleation (particles formed from gases) is not implemented.
- In addition, we neglected interactions of particles with water vapour in the air, for example, in clouds or fog. Similar to the formation of secondary particles, such processes (chemical conversion, agglomeration) are not understood well enough in order to integrate them in an impact assessment model.
- The high resolution of damage to human health due to the number of chemical species, sizes classes, solubility, etc. requires a much larger effort considering data collection in order to inventory PM. Until now, the mass was easily determined by measuring PM₁₀ mass. Now the PM₁₀ has to be distinguished in terms of size, chemistry, and solubility.
- Based on first experience compiling LCI and testing the new impact assessment method, it seems that chemical disclosure of the particle mass is a possible bottleneck for the outcome of the results using the new impact assessment.
- Basically the new impact assessment can serve as a stand-alone tool. However, in case the results shall be merged with other midpoint categories, the impact due to PM assessed with established methods has to be manually deleted and subsequently, when assessed with the new impact assessment, added back to the other midpoint categories. This requires some effort to put into practice.

Based on these last limitations considering the new requirements on LCI data modelling and the effort to use the tool together with other midpoint categories in LCA leads to a couple of potential follow-up projects.

14.3 Follow up projects

Up to now all life cycle inventories are compiled in such a way that they meet the requirements of established impact assessment models. Thus, the benefits of the new impact assessment method can only be harvested if the LCI for PM will be compiled according to the requirements of the new method (size segregated and chemically analysed PM fractions).

A first project in which LCI data are collected according to the new model occurs in the framework program "Aktionsplan Holz". In this program the life cycle inventories for wood combustion processes are updated by the Paul Scherrer Institut. The results of this update will allow for a first real and comprehensive test of the new impact assessment.

A broad use of this method requires basically three steps. Each of these steps reflects a potential follow up project:

1. The elementary flows have to be entered into a LCI database such as ecoinvent. This is in fact not a big effort. However, this makes sense only if
2. All datasets in ecoinvent that contain PM emission are adjusted to the new impact assessment method. This means that about 4000 datasets out of 4500 (ecoinvent version 2.2) would have to be reanalysed according to the requirements of the new impact assessment.
3. The adjustment of the LCI datasets is most likely considered only in case that the new method will be incorporated into an established method like ReCiPe or Ecoindicator 99. These two methods are considered as suitable since they use the same unit for the endpoint (DALY).

14.4 Outlook

In the short term it would be ideal to incorporate the new model into an established impact assessment method, where it replaces the existing impact assessment model for PM. For that purpose, inventories would have to be created which can distinguish PM due to the mentioned characteristics. The collection of such inventories is much more complex than before. Instead of a simple measurement of particulate mass, PM has to be divided into different size classes and it has to be chemically analysed. In the ecoinvent database version 2.2 about 4000 life cycle inventories should then be adapted to the new system.

In the medium term

- the model should be extended to new chemical substances, as long as they are present in the ambient air as particles.
- The model could be adapted to other fields of application such as modelling characterisation of nanoparticles and their damage to human health.
- The model can also be adapted by a new parameterization to other geographical regions by doing a completely new parameterization.

In the long term, the model can and shall be expanded. By the time the literature provides more data to other chemical or physical properties (e.g. reactive oxygen species) it is beneficial to incorporate such properties in addition.

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17 Index of tables

Table 1. Characterisation Factors for PM from 5 different impact assessment methods.....	8
Table 2. Overview over main parameters and processes summarised for 6 fate models, partly applied in LCA impact assessment methods.	22
Table 3. Parameters and processes summarised for the same models as used for fate analysis. x = included in the model.....	24
Table 4. Data points found and number of data points required using power analysis. Blue fields indicate the total number of data points for a single substance. Number of paired data points for the 6 substances are shown in the upper right part and sample size required in order to get statistically significant effects using power analysis are shown in the lower left part.	36
Table 5 Data points found and statistical analysis. Number of data points for the 6 substances (upper right part) and P-values from paired sample t-test (lower left part) without α error correction. Red highlighted numbers refer to statistically significant comparisons ($P < 0.05$) and green highlighted values indicate a tendency to a significance ($P < 0.1$).	37
Table 6. Exposure variability between studies. Differences between studies with respect to exposure concentration, number of days the pollutant was measured and the interquartile increase of the pollutant associated with the human health effect.....	38
Table 7. Mean, confidence intervals (CI), number of days (N) for PM ₁₀ and PNC and the difference between the CI and N for PM ₁₀ and PNC.....	40
Table 8. Number of data points (blue fields), number of paired samples (upper right part) and P-values from paired sample t-test (lower left part).	41
Table 9. Composition of sea salt.	44
Table 10. Mean ambient European PM concentration at three different sites.....	46
Table 11. Overview on the most relevant chemical substances and their contributions.....	47
Table 12. Most important assumptions and limitations for the background concentration.....	47
Table 13. Most important assumptions and limitations for the fate model.....	53
Table 14. Most important assumptions and limitations for the exposure model.	56
Table 15. List of chemical substances and groups of substances used for the chemical description of PM. ..	58
Table 16. Modelling of solubility depending on size and chemistry.....	59
Table 17. Effects of solubility depending on particle size.	60
Table 18. Most important assumptions and limitations for the effect model.....	61
Table 19. DALY`s for EUR-A countries provided by Lopez et al. ¹²⁶	61
Table 20. Most important assumptions and limitations for the effect model.....	62
Table 21. Life cycle inventory for wood combustion using 5 different types of wood.....	70
Table 22. Results in DALY/kWh heat per chemical for each type of wood.....	70
Table 23. Results compared with Ecoindicator 99 (EI99) and ReCiPe.	71
Table 24. Inventory for particulate emissions from Solli et al. for the combustion of 1 kg wood.	71

Table 25. Results in DALY/kWh for the combustion process with two different stoves and a comparison between different impact assessment methods.....	72
Table 26. Mass fractions for the different size fractions from Lamberg ¹²⁹ per mg/MJ heat produced for all combustion devices.	73
Table 27. Mass contribution in % of the chemical substances used in the LCI for all combustion devices.	73
Table 28. Results in DALY/MJ heat produced 6 different devices for wood combustion assessed with 3 different IA methods and the share between different impact assessment methods.	77
Table 29. Results in DALY/kg cut natural stone and a comparison between different impact assessment methods.	79
Table 30. Characterisation of the two types of heating.	80
Table 31. Inventory of the emissions for both combustion processes.	81
Table 32. Impact assessment results for the pellet burning process.....	85
Table 33. Impact assessment results for the light fuel oil burning process.....	86
Table 34. Results in DALY/MJ heat produced by burning pellets or fuel and comparison between different impact assessment methods for PM.....	89
Table 35. Comparison of established and the new impact assessment in an overview over all case studies.	90
Table 36. Economic damage of PM for all case studies as a function of the respective functional unit.	93
Table 37. Economic differences between assessments using the new and established methods.....	94

18 Index of figures

Figure 1. Damage due to PM as a share on total human health assessed with 3 different methods.....	7
Figure 2. Phases of an LCA according to ISO 14040.....	10
Figure 3. Framework for the impact assessment method Ecoindicator 99.	12
Figure 4. Steps required for the calculation of CF for PM emissions.	14
Figure 5. Penetration depth of inhaled particles depending on particle size. ¹⁶	17
Figure 6. An algae cell surrounded by carbon black.	18
Figure 7. Residence time, particle radius and removal processes.....	23
Figure 8. Lung deposition model. ²⁷⁻²⁹	24
Figure 9. Estimates of association with 95% confidence interval for PNC on cardiovascular outcomes.	32
Figure 10. Estimates of association with 95% confidence interval for PNC on respiratory outcomes.	33
Figure 11. Mean of PM size classes and the secondary PM.	36
Figure 12. Removal process depending on particle size - Source: Seinfeld & Pandis ²⁰	48
Figure 13. Results removal processes.....	52
Figure 14. Uptake per respiratory department: AV: alveolar; NP: naso-pharyngeal; TB: trachea-bronchial.	54
Figure 15. Particle removal and particle deposition as a function of particle diameter.	55
Figure 16: Relative risk and Odds ratio (left panel) and after normalisation to PM ₁₀ (right panel).....	57
Figure 17. Model verification for size and solubility.....	64
Figure 18. Model verification for population densities.	65
Figure 19. Model verification stack heights.....	65
Figure 20. CF for all chemicals for PM _{2.5} for the base case.	66
Figure 21. Damage of lung cancer split into the main PM air pollutants	67
Figure 22. Damage of CPD split into the main PM air pollutants.	67
Figure 23. Total damage split into the main PM air pollutants.....	67
Figure 24. Damage accounted for all size fractions resolved in chemical components.	74
Figure 25. Damage accounted for PM ₁₀ resolved in the chemical components	74
Figure 26. Damage accounted for PM _{2.5} resolved in the chemical components	75
Figure 27. Damage accounted for UFP resolved in the chemical components	75
Figure 28. Comparison of the new impact assessment method with EI99 and ReCiPe.	76
Figure 29. Flow diagram for the processing of the manufacture of natural stone plate taken from ecoinvent ¹³⁰	78
Figure 30. Total contribution of all processes assessed with ReCiPe.	82
Figure 31. Total damage depicted with respect to all the impact categories, but for the fuel burning process only.....	82
Figure 32. Size segregated damage on a log scale for the pellet burning process.....	83

Figure 33. Size segregated damage on a normal scale for the pellet burning process.....	84
Figure 34. Size segregated damage on a log scale for light fuel oil burning process.	85
Figure 35. Size segregated damage on a normal scale for the light fuel oil burning process.....	86
Figure 36. Step by step: Original impact of Particles – reduced impact after deleting particulate substance – adding back for both processes.....	87
Figure 37. Total damage depicted with respect to all the impact categories, but for the fuel burning process only.....	88
Figure 38. Total contribution of all processes after reassessment.	88
Figure 39. The figure taken from ExternE depicts the impact pathway analysis ¹³⁴	91

19 Annex

Table A1 lists single data points and confidence intervals the effects of particle size on human health.

	RR PM10	CI Pm10	RR PM2.5	CI PM2.5	RR PNC	CI PNC	RR SOx	CI SOx	RR NOx	CI NOx	RR CO	CI CO	
	PM10		PM2.5		PNC		SOx		NOx		CO		
Anderson	1.030	0.020	1.030	0.025	1.030	0.020			1.000	0.025	1.020	0.025	
	1.060	0.035	1.000	0.050	1.040	0.035			1.060	0.055	1.040	0.050	
	1.020	0.095	1.150	0.160	1.070	0.095			1.040	0.130	1.000	0.119	
Anderson	1.020	0.055			1.080	0.110			1.070	0.080	1.070	0.100	
	1.010	0.065			1.080	0.125			1.080	0.085	1.100	0.110	
Atkinson	1.001	0.008	1.002	0.007	1.022	0.016	1.000	0.010	0.998	0.008			
	1.003	0.005	1.003	0.005	1.009	0.012	1.000	0.006	1.002	0.005			
	1.020	0.012	1.016	0.012	1.012	0.023	1.001	0.016	1.002	0.015			
	1.008	0.010	1.008	0.058	0.985	0.015	1.006	0.010	1.015	0.007			
Beelen			1.040	0.155			0.940	0.120	1.070	0.135			
			1.070	0.385			0.880	0.290	1.370	0.435			
			1.060	0.280			1.000	0.235	0.910	0.215			
Belleudi	0.995	0.017	1.015	0.034	1.006	0.021							
	1.032	0.027	1.019	0.302	1.027	0.024							
	1.021	0.033	1.036	0.047	0.996	0.030							
	0.982	0.028	0.999	0.039	0.999	0.026							
Breitner	1.006	0.026	1.017	0.025	1.055	0.045			1.012	0.034	1.021	0.035	
	1.017	0.027	1.025	0.027	1.063	0.048			1.024	0.036	1.024	0.037	
Dadvand	0.997	0.009					0.967	0.010	1.005	0.004	1.241	0.402	
Forrestriere	1.061	0.057			1.083	0.067			1.029	0.059	1.070	0.065	
Gehring	1.080	0.155							1.170	0.160			
Halonen	1.026	0.028	1.026	0.028	1.061	0.030			1.109	0.046	1.043	0.030	
	1.027	0.025	1.015	0.023	1.025	0.025			1.037	0.036	1.009	0.025	
	1.025	0.023	1.031	0.022	1.024	0.035			1.048	0.036	1.037	0.025	
Halonen	0.993	0.019	1.009	0.018	1.000	0.019							
	1.004	0.050	1.014	0.043	1.033	0.041							
	1.002	0.005	1.008	0.018	1.004	0.018							
	0.978	0.026	0.992	0.024	1.026	0.037							
	0.989	0.026	1.002	0.024	1.041	0.038							
	1.008	0.028	1.031	0.025	1.039	0.038							
	1.027	0.029	1.025	0.026	1.017	0.024							
	1.002	0.039	1.019	0.034	1.077	0.048							
	Jalaludin	1.003	0.009	1.009	0.007			1.006	0.012	1.013	0.010	1.024	0.010
		1.010	0.010	1.013	0.008			1.011	0.015	1.018	0.013	1.029	0.012
1.006		0.016	1.012	0.012			1.003	0.022	1.017	0.018	1.029	0.018	
0.980		0.018	0.989	0.015			0.976	0.024	0.983	0.023	1.003	0.022	
Kettunen	0.994	0.028	1.006	0.036	1.008	0.068			0.991	0.060	1.005	0.038	
	1.109	0.104	1.074	0.063	1.085	0.101			1.039	0.094	1.083	0.080	
Lanki	1.003	0.008			1.005	0.009			1.003	0.008	1.005	0.005	
	1.003	0.009			1.013	0.013			1.005	0.009	1.007	0.006	
Leitte	1.003	0.030			1.030	0.055	1.020	0.035	1.030	0.035			
	1.003	0.040			1.070	0.085	1.040	0.070	1.060	0.060			
	1.020	0.035			1.060	0.050	1.020	0.025	1.010	0.020			
Lipfert Naess	1.072	0.061	1.118	0.083			1.045	0.107	1.051	0.088	1.032	0.082	
	1.020	0.055	1.100	0.055					1.080	0.045			
	1.072	0.180	1.270	0.180					1.210	0.170			
	1.090	0.105	1.070	0.105					1.070	0.105			
	1.110	0.075	1.140	0.075					1.070	0.070			
	1.070	0.150	1.090	0.155					1.060	0.145			
	1.110	0.150	1.270	0.150					1.230	0.140			
Peel	1.003	0.022	1.016	0.019	0.984	0.016	0.998	0.030					
	1.010	0.010					1.009	0.015	1.025	0.013	1.020	0.010	
Pope	1.030	0.070	1.110	0.090			1.090	0.050	1.010	0.040	0.960	0.040	
	1.100	0.120	1.170	0.100			1.090	0.075	1.030	0.060	0.940	0.050	
Rosenlund	1.030	0.135	1.250	0.170			1.080	0.100	0.950	0.070	0.900	0.060	
	1.000	0.240					1.030	0.290	0.990	0.270	1.040	0.160	
	1.057	0.057					1.078	0.069	1.041	0.069			
Samoli Sarnat			1.025	0.014			1.018	0.010	1.018	0.022			
			1.028	0.016			1.014	0.023	1.006	0.013			

Slaughter	1.000	0.030	1.010	0.035								
	1.020	0.030	1.020	0.025							1.030	0.030
Stözl	1.004	0.025	1.007	0.023	1.042	0.028			1.012	0.028	1.013	0.022
	1.007	0.027	1.003	0.022	1.031	0.029						
Tolbert	1.004	0.015	1.005	0.012			0.999	0.012				
	1.007	0.012	1.005	0.010			1.007	0.011				
	1.004	0.012					1.003	0.008	1.015	0.011	1.020	0.010
	0.983	0.009					1.003	0.006	1.015	0.011	1.016	0.007
Villeneuve	1.000	0.045	1.010	0.050			1.060	0.095	1.050	0.065	1.010	0.070
	1.030	0.110	1.070	0.090			1.040	0.125	1.050	0.100	1.000	0.060
	0.990	0.055	0.990	0.045			1.060	0.060	0.990	0.060	1.000	0.045
von Klot	1.026	0.032			1.039	0.042			1.028	0.032	1.022	0.025
	1.008	0.023			1.020	0.028			1.032	0.026	1.009	0.017
	1.021	0.018			1.026	0.022			1.032	0.019	1.014	0.013
max	1.110	0.240	1.270	0.385	1.085	0.125	1.090	0.290	1.370	0.435	1.241	0.402
min	0.978	0.005	0.989	0.005	0.984	0.009	0.880	0.006	0.910	0.004	0.900	0.005
Mean	1.0214	0.0464	1.0462	0.0647	1.0329	0.0402	1.0155	0.0620	1.0416	0.0659	1.0246	0.0531
SD	0.032	0.047	0.067	0.078	0.028	0.028	0.042	0.078	0.069	0.076	0.051	0.070
n	68	68	55	55	40	40	32	32	55	55	36	36

Table A1. Single data points and statistics (minimum value, maximum value, mean, standard deviation and total number of data points) of the measure of association (RR=relative risk, odds ratio) and the confidence interval (CI) for PM₁₀, PM_{2.5}, particle number concentration (PNC), SO_x, NO_x and CO.

Table A2 lists the unit risk factors and the reference concentration taken from the New Jersey Department of Environmental Protection.

	CAS No.	Chemical	URF [(ug/m ³) ⁻¹]	Rfc (ug/m ³)
160	1634044	Methyl tert butyl ether	2.6E-07	3000
163	75092	Methylene chloride	4.7E-07	400
102	140885	Ethyl acrylate	5.0E-07	
210	100425	Styrene	5.7E-07	1000
41	133062	Captan	6.6E-07	
121		Gasoline vapors	1.0E-06	15
38	75252	Bromoform	1.1E-06	
100	106898	Epichlorohydrin	1.2E-06	1
17	62533	Aniline	1.6E-06	1
117	75343	Ethylidene dichloride	1.6E-06	500
153	74873	Methyl chloride	1.8E-06	90
231	79016	Trichloroethylene	2.0E-06	600
1	75070	Acetaldehyde	2.2E-06	9
235	1582098	Trifluralin	2.2E-06	
34	117817	Bis(2-ethylhexyl)phthalate	2.4E-06	
103	100414	Ethylbenzene	2.5E-06	
181	86306	Nitrosodiphenylamine (N-)	2.6E-06	
233	88062	Trichlorophenol (2,4,6-)	3.1E-06	
206	75569	Propylene oxide	3.7E-06	30
82	542756	Dichloropropene (1,3-)	4.0E-06	20
134	67721	Hexachloroethane	4.0E-06	
55	95830	Chloro-o-phenylenediamine (4-)	4.6E-06	
189	87865	Pentachlorophenol	5.1E-06	
217	127184	Tetrachloroethylene	5.9E-06	35
13	107051	Allyl chloride	6.0E-06	1
43	56235	Carbon tetrachloride	6.0E-06	40
182	156105	Nitrosodiphenylamine (p-)	6.3E-06	
74	615054	Diaminoanisole (2,4-)	6.6E-06	
20	140578	Aramite	7.1E-06	
215	630206	Tetrachloroethane (1,1,1,2-)	7.4E-06	
97	123911	Dioxane (1,4-)	7.7E-06	
26	71432	Benzene	7.8E-06	30
239	75014	Vinyl chloride	8.8E-06	100
14	117793	Aminoanthraquinone (2-)	9.4E-06	
204	78875	Propylene dichloride	1.0E-05	4
33	108601	Bis(2-chloroisopropyl)ether	1.0E-05	
222	584849	Toluene diisocyanate (2,4-)	1.1E-05	0.07
223	26471625	Toluene diisocyanate (2,4-/2,6-)	1.1E-05	0.07
224	91087	Toluene diisocyanate (2,6-)	1.1E-05	0.07
78	106467	Dichlorobenzene (1,4-)	1.1E-05	800
146		Lead	1.2E-05	
119	50000	Formaldehyde	1.3E-05	9
115	96457	Ethylene thiourea	1.3E-05	
230	79005	Trichloroethane (1,1,2-)	1.6E-05	
2	60355	Acetamide	2.0E-05	
45	108171262	Chlorinated paraffins	2.0E-05	
127	87683	Hexachlorobutadiene	2.2E-05	
53	67663	Chloroform	2.3E-05	
107	107062	Ethylene dichloride	2.6E-05	400
39	106990	Butadiene (1,3-)	3.0E-05	2
24	103333	Azobenzene	3.1E-05	
51	510156	Chlorobenzilate	3.1E-05	
238	593602	Vinyl bromide	3.2E-05	3
169	91203	Naphthalene	3.4E-05	3
175	98953	Nitrobenzene	4.0E-05	9
18	90040	Anisidine (o-)	4.0E-05	
67	120718	Cresidine (p-)	4.3E-05	
211	96093	Styrene oxide	4.6E-05	
30	100447	Benzyl chloride	4.9E-05	
226	95534	Toluidine (o-)	5.1E-05	
216	79345	Tetrachloroethane (1,1,2,2-)	5.8E-05	
70	135206	Cupferron	6.3E-05	
11	107131	Acrylonitrile	6.8E-05	2
56	95692	Chloro-o-toluidine (p-)	7.7E-05	
83	62737	Dichlorvos	8.3E-05	0.5
114	75218	Ethylene oxide	8.8E-05	30
96	121142	Dinitrotoluene (2,4-)	8.9E-05	
72	72559	DDE	9.7E-05	
73	50293	DDT	9.7E-05	
44	57749	Chlordane	1.0E-04	0.7
9	79061	Acrylamide	1.0E-04	6
196	1336363	Polychlorinated biphenyls (PCBs)	1.0E-04	
199	7758012	Potassium bromate	1.4E-04	
99	122667	Diphenylhydrazine (1,2-)	2.2E-04	
170		Nickel and compounds	2.4E-04	0.05
167	90948	Michler's ketone	2.5E-04	
104	51796	Ethyl carbamate	2.9E-04	
86		Diesel particulate matter	3.0E-04	5
58	126998	Chloroprene	3.0E-04	20

Table A2. List of substances with unit risk factors and reference concentrations

130	58899	Hexachlorocyclohexane (gamma-)	3.1E-04	
227	8001352	Toxaphene	3.2E-04	
81	111444	Dichloroethyl ether	3.3E-04	
79	91941	Dichlorobenzidine (3,3'-)	3.4E-04	
162	101144	Methylene bis(2-chloroaniline) (4,4'-)	4.3E-04	
164	101779	Methylenedianiline (4,4-)	4.6E-04	20
126	118741	Hexachlorobenzene	4.6E-04	
131	608731	Hexachlorocyclohexane (technical grade)	5.1E-04	
129	319857	Hexachlorocyclohexane (beta-)	5.3E-04	
106	106934	Ethylene dibromide	6.0E-04	0.8
188	930552	Nitrosopyrrolidine (N-)	6.1E-04	
65	8007452	Coke oven emissions	6.2E-04	
54	107302	Chloromethyl methyl ether	6.9E-04	
200	1120714	Propane sultone (1,3-)	6.9E-04	
166	60344	Methylhydrazine	1.0E-03	0.02
28	50328	Benzo(a)pyrene	1.1E-03	
225	95807	Toluene-2,4-diamine	1.1E-03	
7	53963	Acetylaminofluorene (2-)	1.3E-03	
91	60117	Dimethylaminoazobenzene (4-)	1.3E-03	
124	76448	Heptachlor	1.3E-03	
179	924163	Nitrosodi-n-butylamine (N-)	1.6E-03	
219	62555	Thioacetamide	1.7E-03	
128	319846	Hexachlorocyclohexane (alpha-)	1.8E-03	
184	59892	Nitrosomorpholine (N-)	1.9E-03	
75	96128	Dibromo-3-chloropropane (1,2-)	2.0E-03	0.2
180	621647	Nitrosodi-n-propylamine (N-)	2.0E-03	
31		Beryllium	2.4E-03	0.02
125	1024573	Heptachlor epoxide	2.6E-03	
176	79469	Nitropropane (2-)	2.7E-03	20
187	100754	Nitrosopiperidine (N-)	2.7E-03	
29	98077	Benzotrichloride	3.7E-03	
92	79447	Dimethylcarbamyl chloride	3.7E-03	
90	77781	Dimethyl sulfate	4.0E-03	
201	57578	Propiolactone (beta-)	4.0E-03	
40		Cadmium	4.2E-03	0.02
76	764410	Dichloro-2-butene (1,4-)	4.2E-03	
21		Arsenic (inorganic)	4.3E-03	0.015
85	60571	Dieldrin	4.6E-03	
137	302012	Hydrazine	4.9E-03	0.2
12	309002	Aldrin	4.9E-03	
138	10034932	Hydrazine sulfate	4.9E-03	
15	92671	Aminobiphenyl (4-)	6.0E-03	
183	10595956	Nitrosomethylethylamine (N-)	6.3E-03	
23	1332214	Asbestos	7.7E-03	
185	759739	Nitroso-n-ethylurea (N-)	7.7E-03	
64		Cobalt	9.0E-03	0.006
61	18540299	Chromium VI (total)	1.2E-02	
178	62759	Nitrosodimethylamine (N-)	1.4E-02	
116	151564	Ethyleneimine	1.9E-02	
186	684935	Nitroso-n-methylurea (N-)	3.4E-02	
177	55185	Nitrosodiethylamine (N-)	4.3E-02	
35	542881	Bis(chloromethyl)ether	6.2E-02	
27	92875	Benzidine	6.7E-02	
95	540738	Dimethylhydrazine (1,2-)	1.6E-01	
133	19408743	Hexachlorodibenzo-p-dioxin, mixture	1.3E+00	
214	1746016	Tetrachlorodibenzo(p)dioxin (2,3,7,8-)	3.7E+01	
94	57147	Dimethylhydrazine (1,1-)		0.002
60		Chromic acid mists (Cr VI)		0.008
62		Chromium VI dissolved aerosols		0.008
135	822060	Hexamethylene diisocyanate		0.01
6	98862	Acetophenone		0.02
8	107028	Acrolein		0.02
49	532274	Chloroacetophenone (2-)		0.03
22	7784421	Arsine		0.05
148		Manganese		0.05
194		Phosphorus (white)		0.07
122	111308	Glutaraldehyde		0.08
63		Chromium VI particulates		0.1
171	1313991	Nickel oxide		0.1
174	88744	Nitroaniline (o-)		0.1
220	7550450	Titanium tetrachloride		0.1
236	1314621	Vanadium or vanadium pentoxide		0.1
19	1309644	Antimony trioxide		0.2
46	7782505	Chlorine		0.2
47	10049044	Chlorine dioxide		0.2
132	77474	Hexachlorocyclopentadiene		0.2
172		Nickel, soluble salts		0.2
149		Mercury (elemental)		0.3
191	75445	Phosgene		0.3
192	7803512	Phosphine		0.3

Table A2. List of substances with unit risk factors and reference concentrations (continued)

32	95524	Biphenyl (1,1-)	0.4
57	76062	Chloropicrin	0.4
165	101688	Methylenediphenyl diisocyanate (4,4'-)	0.6
37	7637072	Boron trifluoride	0.7
147	108316	Maleic anhydride	0.7
150	126987	Methacrylonitrile	0.7
140	74908	Hydrogen cyanide	0.8
10	79107	Acrylic acid	1
123	765344	Glycidaldehyde	1
157	624839	Methyl isocyanate	1
213	7664939	Sulfuric acid	1
143	7783064	Hydrogen sulfide	2
87	111422	Diethanolamine	3
208	7631869	Silica (crystalline, respirable)	3
229	120821	Trichlorobenzene (1,2,4-)	4
152	74839	Methyl bromide	5
84	77736	Dicyclopentadiene	7
234	121448	Triethylamine	7
202	123386	Propionaldehyde	8
193	7664382	Phosphoric acid	10
118	16984488	Fluoride	13
141	7664393	Hydrogen fluoride	14
36	7440428	Boron (elemental)	20
88	112345	Diethylene glycol monobutyl ether	20
101	106887	Epoxybutane (1,2-)	20
112	109864	Ethylene glycol monomethyl ether	20
139	7647010	Hydrogen chloride	20
195	85449	Phthalic anhydride	20
207		Selenium and compounds	20
168		Mineral fibers (<1% free silica)	24
93	68122	Dimethylformamide (N,N-)	30
159	25013154	Methyl styrene (mixed isomers)	40
120	98011	Furfural	50
4	75865	Acetone cyanohydrin	60
5	75058	Acetonitrile	60
113	110496	Ethylene glycol monomethyl ether acetate	90
16	7664417	Ammonia	100
59	75296	Chloropropane (2-)	100
241		Xylene (m-,o-,p-, or mixed isomers)	100
77	95501	Dichlorobenzene (1,2-)	200
80	75718	Dichlorodifluoromethane	200
110	110805	Ethylene glycol monoethyl ether	200
190	108952	Phenol	200
237	108054	Vinyl acetate	200
240	75354	Vinylidene chloride	200
69	98828	Cumene	400
108	107211	Ethylene glycol	400
68		Cresol mixtures	600
42	75150	Carbon disulfide	700
136	110543	Hexane (N-)	700
158	80626	Methyl methacrylate	700
232	75694	Trichlorofluoromethane	700
50	108907	Chlorobenzene	1000
154	71556	Methyl chloroform	1000
109	111762	Ethylene glycol monobutyl ether	1600
144	78591	Isophorone	2000
205	107982	Propylene glycol monomethyl ether	2000
161	108872	Methylcyclohexane	3000
203	115071	Propylene	3000
151	67561	Methanol	4000
155	78933	Methyl ethyl ketone	5000
221	108883	Toluene	5000
228	76131	Trichloro-1,2,2-trifluoroethane (1,1,2-)	30000
3	67641	Acetone	31000
89	75376	Difluoroethane (1,1-)	40000
48	75683	Chloro-1,1-difluoroethane (1-) (HCFC-142b)	50000
52	75456	Chlorodifluoromethane (HCFC-22)	50000
218	811972	Tetrafluoroethane (1,1,1,2-)	80000
25		Barium	
66		Copper	
71	110827	Cyclohexane	
105	75003	Ethyl chloride	
111	111159	Ethylene glycol monoethyl ether acetate	
142	7783075	Hydrogen selenide	
145	67630	Isopropanol	
156	108101	Methyl isobutyl ketone	
173	7697372	Nitric acid	
209	1310732	Sodium hydroxide	
212		Sulfates	

Table A2. List of substances with unit risk factors and reference concentrations (continued)

Table A3 lists size segregated concentration of chemical substances with respect to different emission sites.

Ambient air concentration of chemical substances

Pollutant unit site	PM10 µg/m3			PM2.5 µg/m3			UFP Particles/cm3		
	rural	urban	kerbside	rural	urban	kerbside	rural	urban	kerbside
	Black Carbon	9.8E-01	1.7E+00	5.4E+00	8.9E-01	1.7E+00	4.1E+00	2.7E+02	7.2E+02
Nitrate	1.1E+00	5.1E+00	4.2E+00	5.5E-01	3.6E+00	2.6E+00	1.9E+02	4.9E+02	9.2E+02
Ammonia	9.8E-01	2.4E+00	1.7E+00	8.9E-01	2.1E+00	1.7E+00	1.3E+02	3.6E+02	6.7E+02
Sulfates	3.1E+00	4.4E+00	4.2E+00	3.2E+00	3.6E+00	3.1E+00	1.9E+02	5.2E+02	9.6E+02
Asbestos	6.1E-08	1.3E-07	1.7E-07	6.5E-08	1.3E-07	1.5E-07	2.6E-05	7.0E-05	1.3E-04
Sea salt total	1.3E+00	1.4E+00	1.3E+00	3.3E-01	6.4E-01	2.4E-01	1.4E+02	3.7E+02	6.8E+02
Chloride (Cl-)	7.2E-01	7.5E-01	6.9E-01	1.8E-01	3.5E-01	1.3E-01	7.5E+01	2.0E+02	3.8E+02
Sodium (Na+)	4.0E-01	4.2E-01	3.8E-01	1.0E-01	2.0E-01	7.3E-02	4.2E+01	1.1E+02	2.1E+02
Sulfate (SO42-)	1.0E-01	1.0E-01	9.6E-02	2.5E-02	4.9E-02	1.8E-02	1.1E+01	2.8E+01	5.2E+01
Magnesium (Mg2+)	4.8E-02	5.0E-02	4.6E-02	1.2E-02	2.4E-02	8.8E-03	5.0E+00	1.3E+01	2.5E+01
Calcium (Ca2+)	1.5E-02	1.6E-02	1.5E-02	3.9E-03	7.5E-03	2.8E-03	1.6E+00	4.3E+00	8.1E+00
Potassium (K+)	1.5E-02	1.5E-02	1.4E-02	3.7E-03	7.1E-03	2.7E-03	1.5E+00	4.1E+00	7.6E+00
Bicarbonate (HCO3-)	5.4E-03	5.6E-03	5.1E-03	1.4E-03	2.6E-03	9.8E-04	5.6E-01	1.5E+00	2.8E+00
Bromide (Br-)	2.5E-03	2.6E-03	2.4E-03	6.3E-04	1.2E-03	4.5E-04	2.6E-01	6.9E-01	1.3E+00
Borate (BO33-)	1.0E-03	1.1E-03	1.0E-03	2.7E-04	5.1E-04	1.9E-04	1.1E-01	2.9E-01	5.5E-01
Strontium (Sr2+)	5.2E-04	5.4E-04	5.0E-04	1.3E-04	2.6E-04	9.6E-05	5.5E-02	1.5E-01	2.7E-01
Misconstituents	1.3E-04	1.4E-04	1.3E-04	3.3E-05	6.4E-05	2.4E-05	1.4E-02	3.7E-02	6.8E-02
Mineral dust total	9.4E-01	1.7E+00	4.5E+00	3.2E-01	8.5E-01	1.1E+00	4.1E+02	1.1E+03	2.0E+03
SiO2	1.3E-01	2.5E-01	6.5E-01	4.5E-02	1.2E-01	1.6E-01	5.8E+01	1.6E+02	2.9E+02
Al2O3	1.3E-01	2.5E-01	6.5E-01	4.5E-02	1.2E-01	1.6E-01	5.8E+01	1.6E+02	2.9E+02
CaCO3	1.3E-01	2.5E-01	6.5E-01	4.5E-02	1.2E-01	1.6E-01	5.8E+01	1.6E+02	2.9E+02
MgCO3	1.3E-01	2.5E-01	6.5E-01	4.5E-02	1.2E-01	1.6E-01	5.8E+01	1.6E+02	2.9E+02
FeO	1.3E-01	2.5E-01	6.5E-01	4.5E-02	1.2E-01	1.6E-01	5.8E+01	1.6E+02	2.9E+02
Fe2O3	1.3E-01	2.5E-01	6.5E-01	4.5E-02	1.2E-01	1.6E-01	5.8E+01	1.6E+02	2.9E+02
CaO	1.3E-01	2.5E-01	6.5E-01	4.5E-02	1.2E-01	1.6E-01	5.8E+01	1.6E+02	2.9E+02
Trace elements total	7.0E-01	1.3E+00	3.4E+00	2.4E-01	6.4E-01	8.2E-01	3.1E+02	8.2E+02	1.5E+03
Sodium	8.8E-02	1.6E-01	4.2E-01	3.0E-02	8.0E-02	1.0E-01	3.8E+01	1.0E+02	1.9E+02
Magnesium	1.7E-02	3.1E-02	8.1E-02	5.6E-03	1.5E-02	1.9E-02	7.3E+00	1.9E+01	3.6E+01
Aluminium	1.9E-02	3.5E-02	9.1E-02	6.4E-03	1.7E-02	2.2E-02	8.2E+00	2.2E+01	4.1E+01
Potassium	7.4E-02	1.4E-01	3.6E-01	2.5E-02	6.8E-02	8.7E-02	3.3E+01	8.7E+01	1.6E+02
Calcium	5.7E-02	1.1E-01	2.7E-01	1.9E-02	5.2E-02	6.6E-02	2.5E+01	6.6E+01	1.2E+02
Vanadium or V. penta	7.1E-04	1.3E-03	3.4E-03	2.4E-04	6.5E-04	8.3E-04	3.1E-01	8.3E-01	1.5E+00
Manganese	1.9E-03	3.6E-03	9.4E-03	6.6E-04	1.8E-03	2.3E-03	8.5E-01	2.3E+00	4.2E+00
Iron	1.1E-01	2.1E-01	5.3E-01	3.7E-02	1.0E-01	1.3E-01	4.8E+01	1.3E+02	2.4E+02
Nickel and compound	6.2E-04	1.2E-03	3.0E-03	2.1E-04	5.6E-04	7.2E-04	2.7E-01	7.2E-01	1.3E+00
Copper	4.3E-03	8.1E-03	2.1E-02	1.5E-03	3.9E-03	5.0E-03	1.9E+00	5.0E+00	9.4E+00
Galium	2.2E-05	4.0E-05	1.0E-04	7.3E-06	2.0E-05	2.5E-05	9.5E-03	2.5E-02	4.7E-02
Arsenic (inorganic)	1.7E-04	3.1E-04	8.0E-04	5.6E-05	1.5E-04	1.9E-04	7.2E-02	1.9E-01	3.6E-01
Selenium and compo	1.5E-04	2.8E-04	7.2E-04	5.1E-05	1.4E-04	1.7E-04	6.5E-02	1.7E-01	3.3E-01
Rubidium	1.8E-04	3.4E-04	8.9E-04	6.2E-05	1.7E-04	2.1E-04	8.0E-02	2.1E-01	4.0E-01
Yttrium	1.5E-05	2.9E-05	7.5E-05	5.2E-06	1.4E-05	1.8E-05	6.8E-03	1.8E-02	3.4E-02
Molybdenum	3.6E-04	6.8E-04	1.8E-03	1.2E-04	3.3E-04	4.2E-04	1.6E-01	4.2E-01	7.9E-01
Rhodium	1.1E-06	2.0E-06	5.3E-06	3.7E-07	9.9E-07	1.3E-06	4.8E-04	1.3E-03	2.4E-03
Cadmium	2.1E-04	4.0E-04	1.0E-03	7.2E-05	2.0E-04	2.5E-04	9.4E-02	2.5E-01	4.7E-01
Antimony	4.0E-04	7.4E-04	1.9E-03	1.3E-04	3.6E-04	4.7E-04	1.7E-01	4.6E-01	8.7E-01
Lanthanum	1.7E-04	3.2E-04	8.2E-04	5.8E-05	1.6E-04	2.0E-04	7.4E-02	2.0E-01	3.7E-01
Neodymium	2.6E-05	4.9E-05	1.3E-04	8.9E-06	2.4E-05	3.1E-05	1.1E-02	3.1E-02	5.7E-02
Thallium	1.3E-05	2.4E-05	6.3E-05	4.4E-06	1.2E-05	1.5E-05	5.7E-03	1.5E-02	2.8E-02
Lead	5.4E-03	1.0E-02	2.6E-02	1.8E-03	4.9E-03	6.3E-03	2.3E+00	6.2E+00	1.2E+01
Silicon	3.9E-02	7.2E-02	1.9E-01	1.3E-02	3.5E-02	4.5E-02	1.7E+01	4.5E+01	8.4E+01
Titanium	6.0E-03	1.1E-02	2.9E-02	2.0E-03	5.4E-03	7.0E-03	2.6E+00	6.9E+00	1.3E+01
Chromium VI	4.4E-03	8.2E-03	2.1E-02	1.5E-03	4.0E-03	5.2E-03	1.9E+00	5.1E+00	9.6E+00

Cobalt	7.3E-04	1.4E-03	3.6E-03	2.5E-04	6.7E-04	8.6E-04	3.2E-01	8.5E-01	1.6E+00
Scandium	2.7E-04	5.0E-04	1.3E-03	9.0E-05	2.4E-04	3.1E-04	1.2E-01	3.1E-01	5.8E-01
Zinc	8.6E-03	1.6E-02	4.1E-02	2.9E-03	7.8E-03	1.0E-02	3.7E+00	1.0E+01	1.9E+01
Bromine	1.2E-03	2.2E-03	5.6E-03	3.9E-04	1.1E-03	1.4E-03	5.1E-01	1.4E+00	2.5E+00
Sulfur	2.6E-01	4.8E-01	1.3E+00	8.7E-02	2.4E-01	3.0E-01	1.1E+02	3.0E+02	5.6E+02
Barium	4.1E-03	7.7E-03	2.0E-02	1.4E-03	3.7E-03	4.8E-03	1.8E+00	4.8E+00	8.9E+00
Cesium	6.7E-05	1.2E-04	3.2E-04	2.2E-05	6.1E-05	7.8E-05	2.9E-02	7.7E-02	1.4E-01
Gold	1.4E-06	2.5E-06	6.6E-06	4.6E-07	1.2E-06	1.6E-06	6.0E-04	1.6E-03	3.0E-03
Mercury (elemental)	4.2E-05	7.9E-05	2.1E-04	1.4E-05	3.9E-05	5.0E-05	1.9E-02	4.9E-02	9.2E-02
Cerium	7.0E-04	1.3E-03	3.4E-03	2.4E-04	6.4E-04	8.2E-04	3.1E-01	8.2E-01	1.5E+00
Samarium	2.5E-05	4.7E-05	1.2E-04	8.5E-06	2.3E-05	2.9E-05	1.1E-02	2.9E-02	5.5E-02
Europium	9.6E-05	1.8E-04	4.6E-04	3.2E-05	8.7E-05	1.1E-04	4.2E-02	1.1E-01	2.1E-01
Ytterbium	9.4E-05	1.8E-04	4.6E-04	3.2E-05	8.6E-05	1.1E-04	4.1E-02	1.1E-01	2.0E-01
Lutetium	5.4E-06	1.0E-05	2.6E-05	1.8E-06	5.0E-06	6.4E-06	2.4E-03	6.3E-03	1.2E-02
Thorium	2.5E-05	4.7E-05	1.2E-04	8.6E-06	2.3E-05	3.0E-05	1.1E-02	3.0E-02	5.5E-02
Uranium	1.5E-05	2.8E-05	7.3E-05	5.1E-06	1.4E-05	1.8E-05	6.6E-03	1.8E-02	3.3E-02
Organic matter total	2.3E-04	3.9E-04	5.3E-04	1.9E-04	4.1E-04	3.6E-04	5.2E-01	1.4E+00	2.6E+00
Dioxins, PCDD	1.2E-05	2.1E-05	2.8E-05	1.0E-05	2.2E-05	1.9E-05	2.8E-02	7.4E-02	1.4E-01
Furane, PCDF	1.6E-05	2.8E-05	3.8E-05	1.3E-05	2.9E-05	2.6E-05	3.7E-02	9.9E-02	1.9E-01
PCB	2.0E-04	3.4E-04	4.7E-04	1.7E-04	3.6E-04	3.2E-04	4.6E-01	1.2E+00	2.3E+00
aliphatic total	2.5E+00	6.4E+00	8.6E+00	2.4E+00	4.4E+00	6.5E+00	1.8E+03	4.9E+03	9.1E+03
Alkanes, -enes, -ines	5.0E-01	1.3E+00	1.7E+00	4.8E-01	8.9E-01	1.3E+00	3.7E+02	9.9E+02	1.9E+03
Aldehydes	7.6E-02	2.0E-01	2.7E-01	7.4E-02	1.4E-01	2.0E-01	5.7E+01	1.5E+02	2.8E+02
Ketones	7.6E-02	2.0E-01	2.7E-01	7.4E-02	1.4E-01	2.0E-01	5.7E+01	1.5E+02	2.8E+02
Alkanol	4.3E-01	1.1E+00	1.5E+00	4.2E-01	7.7E-01	1.1E+00	3.2E+02	8.6E+02	1.6E+03
Organic acid	1.4E+00	3.6E+00	4.8E+00	1.3E+00	2.5E+00	3.6E+00	1.0E+03	2.7E+03	5.1E+03
Aromatics total	1.7E-01	4.4E-01	5.9E-01	1.6E-01	3.0E-01	4.5E-01	1.3E+02	3.4E+02	6.3E+02
Aromatics (monocyclic)	1.2E-01	3.2E-01	4.4E-01	1.2E-01	2.2E-01	3.3E-01	9.3E+01	2.5E+02	4.6E+02
PAH	4.5E-02	1.2E-01	1.6E-01	4.4E-02	8.0E-02	1.2E-01	3.4E+01	9.0E+01	1.7E+02

Table A3. Detailed list of all chemically identified substances for this study subject to site specification.

Figure A1 depicts size dependent coagulation coefficients for Brownian coagulation.

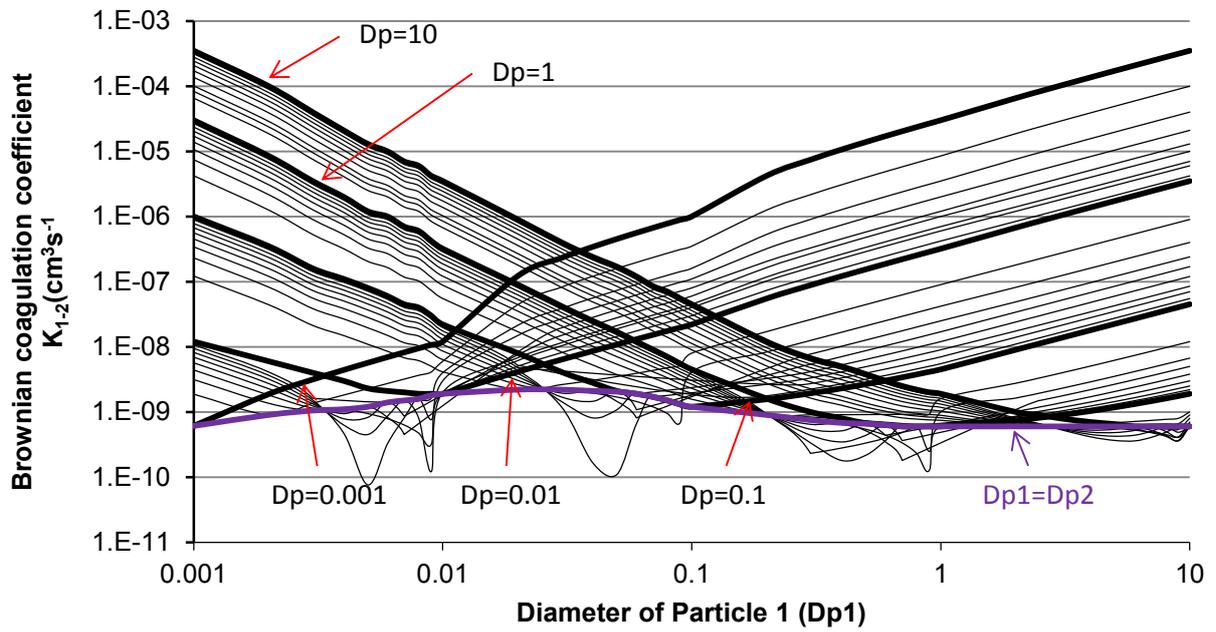


Figure A1. Brownian coagulation coefficient $K_{1,2}$ for coagulation in air at 25°C of particles of diameters D_{p1} and D_{p2} . To use this figure, find the smaller of the two particles as the abscissa and then locate the line corresponding to the larger particle.

Table A4 lists the 93 characterisation factors derived for the base case (unspecific solubility, low stack height, high population density, urban site). The name is compiled in a way that it depicts unambiguously all parameters included in the model. Each parameter is separated with a diagonal slash:

“size”/“solubility”/“population density_location site”/“stack height”/“chemical substance”

Name	DALY/kg,1 Partikel	Nr.
PM10/unspecific/high pop.density_urban/low stack/Black Carbon	4.18E-09	1
PM10/unspecific/high pop.density_urban/low stack/Nitrate	4.18E-09	2
PM10/unspecific/high pop.density_urban/low stack/Ammonia	3.39E-06	3
PM10/unspecific/high pop.density_urban/low stack/Sulfates	4.18E-09	4
PM10/unspecific/high pop.density_urban/low stack/Barium	4.18E-09	5
PM10/unspecific/high pop.density_urban/low stack/Asbestos	6.11E-05	6
PM10/unspecific/high pop.density_urban/low stack/Mercury (elemental)	1.13E-03	7
PM10/unspecific/high pop.density_urban/low stack/Chromium VI	4.34E-03	8
PM10/unspecific/high pop.density_urban/low stack/Vanadium or vanadium pentoxide	3.39E-03	9
PM10/unspecific/high pop.density_urban/low stack/Manganese	6.77E-03	10
PM10/unspecific/high pop.density_urban/low stack/Copper	4.18E-09	11
PM10/unspecific/high pop.density_urban/low stack/Lead	9.85E-08	12
PM10/unspecific/high pop.density_urban/low stack/Nickel and compounds	6.77E-03	13
PM10/unspecific/high pop.density_urban/low stack/Cadmium	1.70E-02	14
PM10/unspecific/high pop.density_urban/low stack/Cobalt	5.65E-02	15
PM10/unspecific/high pop.density_urban/low stack/Inorganic salt	4.18E-09	16
PM10/unspecific/high pop.density_urban/low stack/Iron	4.18E-09	17
PM10/unspecific/high pop.density_urban/low stack/Selenium and compounds	1.69E-05	18
PM10/unspecific/high pop.density_urban/low stack/Arsenic (inorganic)	2.26E-02	19
PM10/unspecific/high pop.density_urban/low stack/Alkanes/Alkenes/Alkines	3.40E-05	20
PM10/unspecific/high pop.density_urban/low stack/Aldehydes	3.08E-05	21
PM10/unspecific/high pop.density_urban/low stack/Ketones	5.47E-06	22
PM10/unspecific/high pop.density_urban/low stack/Alkanol	6.37E-07	23
PM10/unspecific/high pop.density_urban/low stack/Organic Acid	3.07E-05	24
PM10/unspecific/high pop.density_urban/low stack/Aromatics (monocyclic and polycyclic with heteroatoms)	1.37E-05	25
PM10/unspecific/high pop.density_urban/low stack/PAH	8.73E-06	26
PM10/unspecific/high pop.density_urban/low stack/PCB	7.96E-07	27
PM10/unspecific/high pop.density_urban/low stack/Dioxins, PCDD	2.93E-01	28
PM10/unspecific/high pop.density_urban/low stack/Furane, PCDF	2.93E-02	29
PM10/unspecific/high pop.density_urban/low stack/Zinc	4.18E-09	30
PM10/unspecific/high pop.density_urban/low stack/Mineral dust	2.39E-09	31
PM2.5/unspecific/high pop.density_urban/low stack/Black Carbon	6.74E-09	32
PM2.5/unspecific/high pop.density_urban/low stack/Nitrate	9.52E-09	33
PM2.5/unspecific/high pop.density_urban/low stack/Ammonia	6.12E-06	34
PM2.5/unspecific/high pop.density_urban/low stack/Sulfates	8.25E-09	35
PM2.5/unspecific/high pop.density_urban/low stack/Barium	1.39E-08	36
PM2.5/unspecific/high pop.density_urban/low stack/Asbestos	9.71E-05	37
PM2.5/unspecific/high pop.density_urban/low stack/Mercury (elemental)	3.75E-03	38
PM2.5/unspecific/high pop.density_urban/low stack/Chromium VI	1.44E-02	39
PM2.5/unspecific/high pop.density_urban/low stack/Vanadium or vanadium pentoxide	1.12E-02	40
PM2.5/unspecific/high pop.density_urban/low stack/Manganese	2.25E-02	41
PM2.5/unspecific/high pop.density_urban/low stack/Copper	1.39E-08	42
PM2.5/unspecific/high pop.density_urban/low stack/Lead	3.27E-07	43
PM2.5/unspecific/high pop.density_urban/low stack/Nickel and compounds	2.25E-02	44
PM2.5/unspecific/high pop.density_urban/low stack/Cadmium	5.63E-02	45
PM2.5/unspecific/high pop.density_urban/low stack/Cobalt	1.88E-01	46

Table A4. The CF`s for UFP refer to damage in DALY per 1 particle of the specific substance while the CF`s for PM₁₀ and PM_{2.5} refer to damage in DALY per 1 kg of the specific substance.

Name	DALY/kg,1 Partikel	Nr.
PM2.5/unspecific/high pop.density_urban/low stack/Inorganic salt	1.44E-08	47
PM2.5/unspecific/high pop.density_urban/low stack/Iron	1.39E-08	48
PM2.5/unspecific/high pop.density_urban/low stack/Selenium and compounds	5.62E-05	49
PM2.5/unspecific/high pop.density_urban/low stack/Arsenic (inorganic)	7.50E-02	50
PM2.5/unspecific/high pop.density_urban/low stack/Alkanes/Alkenes/Alkines	7.99E-05	51
PM2.5/unspecific/high pop.density_urban/low stack/Aldehydes	7.22E-05	52
PM2.5/unspecific/high pop.density_urban/low stack/Ketones	1.28E-05	53
PM2.5/unspecific/high pop.density_urban/low stack/Alkanol	1.49E-06	54
PM2.5/unspecific/high pop.density_urban/low stack/Organic Acid	7.22E-05	55
PM2.5/unspecific/high pop.density_urban/low stack/Aromatics (monocyclic and polycyclic with heteroatoms)	3.21E-05	56
PM2.5/unspecific/high pop.density_urban/low stack/PAH	2.05E-05	57
PM2.5/unspecific/high pop.density_urban/low stack/PCB	1.22E-06	58
PM2.5/unspecific/high pop.density_urban/low stack/Dioxins, PCDD	4.51E-01	59
PM2.5/unspecific/high pop.density_urban/low stack/Furane, PCDF	4.51E-02	60
PM2.5/unspecific/high pop.density_urban/low stack/Zinc	1.39E-08	61
PM2.5/unspecific/high pop.density_urban/low stack/Mineral dust	7.92E-09	62
UFP/unspecific/high pop.density_urban/low stack/Black Carbon	2.10E-26	63
UFP/unspecific/high pop.density_urban/low stack/Nitrate	9.22E-26	64
UFP/unspecific/high pop.density_urban/low stack/Ammonia	4.83E-23	65
UFP/unspecific/high pop.density_urban/low stack/Sulfates	7.66E-26	66
UFP/unspecific/high pop.density_urban/low stack/Barium	1.43E-26	67
UFP/unspecific/high pop.density_urban/low stack/Asbestos	2.37E-22	68
UFP/unspecific/high pop.density_urban/low stack/Mercury (elemental)	3.87E-21	69
UFP/unspecific/high pop.density_urban/low stack/Chromium VI	1.49E-20	70
UFP/unspecific/high pop.density_urban/low stack/Vanadium or vanadium pentoxide	1.16E-20	71
UFP/unspecific/high pop.density_urban/low stack/Manganese	2.32E-20	72
UFP/unspecific/high pop.density_urban/low stack/Copper	1.43E-26	73
UFP/unspecific/high pop.density_urban/low stack/Lead	3.38E-25	74
UFP/unspecific/high pop.density_urban/low stack/Nickel and compounds	2.32E-20	75
UFP/unspecific/high pop.density_urban/low stack/Cadmium	5.82E-20	76
UFP/unspecific/high pop.density_urban/low stack/Cobalt	1.94E-19	77
UFP/unspecific/high pop.density_urban/low stack/Inorganic salt	3.33E-26	78
UFP/unspecific/high pop.density_urban/low stack/Iron	1.43E-26	79
UFP/unspecific/high pop.density_urban/low stack/Selenium and compounds	5.81E-23	80
UFP/unspecific/high pop.density_urban/low stack/Arsenic (inorganic)	7.76E-20	81
UFP/unspecific/high pop.density_urban/low stack/Alkanes/Alkenes/Alkines	9.46E-23	82
UFP/unspecific/high pop.density_urban/low stack/Aldehydes	8.55E-23	83
UFP/unspecific/high pop.density_urban/low stack/Ketones	1.52E-23	84
UFP/unspecific/high pop.density_urban/low stack/Alkanol	1.77E-24	85
UFP/unspecific/high pop.density_urban/low stack/Organic Acid	8.54E-23	86
UFP/unspecific/high pop.density_urban/low stack/Aromatics (monocyclic and polycyclic with heteroatoms)	3.80E-23	87
UFP/unspecific/high pop.density_urban/low stack/PAH	2.42E-23	88
UFP/unspecific/high pop.density_urban/low stack/PCB	4.72E-25	89
UFP/unspecific/high pop.density_urban/low stack/Dioxins, PCDD	1.74E-19	90
UFP/unspecific/high pop.density_urban/low stack/Furane, PCDF	1.74E-20	91
UFP/unspecific/high pop.density_urban/low stack/Zinc	1.43E-26	92
UFP/unspecific/high pop.density_urban/low stack/Mineral dust	8.19E-27	93

Table A4. The CF's for UFP refer to damage in DALY per 1 particle of the specific substance while the CF's for PM₁₀ and PM_{2.5} refer to damage in DALY per 1 kg of the specific substance (continued).

Table A5 lists all damage values split in size and chemical substance in DALY/MJ heat for case study 3.

PM10	Device 1	Device 2	Device 3	Device 4	Device 5	Device 6
CB	5.54E-14	8.71E-13	1.20E-13	1.05E-13	3.61E-16	2.45E-13
OM	5.60E-11	7.45E-10	3.31E-11	3.47E-11	7.16E-12	6.64E-10
Cd	2.06E-11	1.30E-10	3.14E-11	5.50E-11	2.49E-12	1.45E-11
Co	9.99E-11	1.32E-09	8.12E-11	8.27E-10	4.88E-12	2.54E-12
Cr	1.35E-11	1.59E-12	1.48E-12	1.51E-12	4.97E-12	9.73E-12
Cu	1.36E-17	3.02E-17	8.58E-18	2.40E-17	2.34E-17	1.88E-19
Mn	2.77E-11	1.24E-09	1.32E-11	1.91E-11	1.17E-10	3.05E-13
Ni	3.03E-12	4.61E-11	4.87E-12	4.39E-12	5.84E-13	3.05E-13
Pb	6.54E-16	1.13E-15	7.29E-16	1.13E-15	3.32E-16	2.80E-16
V	1.87E-13	1.58E-12	9.04E-14	7.43E-14	2.92E-13	1.52E-13
As	1.25E-12	9.62E-12	8.59E-13	2.36E-12	8.78E-13	1.02E-12
Fe	4.85E-17	2.84E-15	1.03E-16	8.74E-17	5.05E-18	1.88E-19
Zn	1.46E-15	4.98E-15	3.13E-15	5.68E-15	3.03E-16	1.82E-15
PAH	4.09E-12	3.50E-11	4.75E-12	3.01E-13	3.88E-16	3.35E-11
unspecified	3.24E-10	-1.36E-10	2.53E-10	2.97E-10	2.10E-10	2.50E-10
PM2.5	Device 1	Device 2	Device 3	Device 4	Device 5	Device 6
CB	6.90E-13	2.65E-12	1.04E-12	8.73E-13	2.79E-15	5.44E-12
OM	1.01E-09	3.29E-09	4.18E-10	4.20E-10	8.06E-11	2.15E-08
Cd	5.28E-10	8.12E-10	5.59E-10	9.42E-10	3.96E-11	6.64E-10
Co	2.56E-09	8.27E-09	1.45E-09	1.42E-08	7.76E-11	1.16E-10
Cr	1.04E-10	3.00E-12	7.94E-12	7.77E-12	2.38E-11	1.34E-10
Cu	3.49E-16	1.89E-16	1.53E-16	4.12E-16	3.73E-16	8.61E-18
Mn	7.09E-10	7.75E-09	2.36E-10	3.27E-10	1.86E-09	1.40E-11
Ni	7.76E-11	2.88E-10	8.68E-11	7.52E-11	9.30E-12	1.40E-11
Pb	1.67E-14	7.08E-15	1.30E-14	1.94E-14	5.28E-15	1.28E-14
V	4.79E-12	9.91E-12	1.61E-12	1.27E-12	4.65E-12	6.98E-12
As	3.20E-11	6.02E-11	1.53E-11	4.05E-11	1.40E-11	4.66E-11
Fe	1.24E-15	1.78E-14	1.84E-15	1.50E-15	8.04E-17	8.61E-18
Zn	3.73E-14	3.11E-14	5.58E-14	9.73E-14	4.82E-15	8.35E-14
PAH	7.40E-11	1.55E-10	5.99E-11	3.64E-12	4.37E-15	1.08E-09
unspecified	5.67E-09	-5.80E-10	3.07E-09	3.47E-09	2.28E-09	7.82E-09
UFP	Device 1	Device 2	Device 3	Device 4	Device 5	Device 6
CB	4.63E-12	1.40E-12	2.99E-12	5.42E-12	3.32E-14	3.78E-12
OM	2.59E-09	6.59E-10	4.56E-10	9.90E-10	3.64E-10	5.66E-09
Cd	1.18E-09	1.42E-10	5.33E-10	1.94E-09	1.56E-10	1.53E-10
Co	5.70E-09	1.44E-09	1.38E-09	2.91E-08	3.06E-10	2.68E-11
Cr	2.32E-10	5.24E-13	7.57E-12	1.60E-11	9.40E-11	3.08E-11
Cu	7.77E-16	3.30E-17	1.46E-16	8.47E-16	1.47E-15	1.98E-18
Mn	1.58E-09	1.35E-09	2.25E-10	6.74E-10	7.34E-09	3.21E-12
Ni	1.73E-10	5.04E-11	8.28E-11	1.55E-10	3.67E-11	3.21E-12
Pb	3.73E-14	1.24E-15	1.24E-14	3.99E-14	2.08E-14	2.94E-15
V	1.07E-11	1.73E-12	1.54E-12	2.62E-12	1.83E-11	1.60E-12
As	7.12E-11	1.05E-11	1.46E-11	8.33E-11	5.51E-11	1.07E-11
Fe	2.76E-15	3.11E-15	1.75E-15	3.08E-15	3.17E-16	1.98E-18
Zn	8.29E-14	5.44E-15	5.32E-14	2.00E-13	1.90E-14	1.92E-14
PAH	1.89E-10	3.10E-11	6.54E-11	8.58E-12	1.97E-14	2.86E-10
unspecified	2.98E-08	-2.39E-10	6.92E-09	1.69E-08	2.12E-08	4.25E-09

Table A5: Case study 3: Detailed results from the impact assessment in DALY/MJ heat.

Table A6 lists all emissions to air for the pellets burning process used in case study 5. Substances labelled with “P” are assumed to be particles.

Acetaldehyde		0.000000061 kg
Ammonia	P	0.00000173 kg
Arsenic	P	0.000000001 kg
Benzene		0.000000091 kg
Benzene, ethyl-		0.000000003 kg
Benzene, hexachloro-		7.2E-15 kg
Benzo(a)pyrene	P	5E-10 kg
Bromine	P	0.000000006 kg
Cadmium	P	7E-10 kg
Calcium	P	0.00000585 kg
Carbon dioxide, biogenic		0.0965 kg
Carbon monoxide, biogenic		0.000096 kg
Chlorine		0.00000018 kg
Chromium	P	3.96E-09 kg
Chromium VI	P	4E-11 kg
Copper	P	0.000000022 kg
Dinitrogen monoxide		0.000003 kg
Dioxin, 2,3,7,8 Tetrachlorodibenzo-p-	P	3.1E-14 kg
Fluorine		0.00000005 kg
Formaldehyde		0.00000013 kg
Heat, waste		1.08 MJ
Hydrocarbons, aliphatic, alkanes, unspecified	P	0.000000091 kg
Hydrocarbons, aliphatic, unsaturated	P	0.00000031 kg
Lead	P	0.000000025 kg
Magnesium	P	0.00000036 kg
Manganese	P	0.00000017 kg
Mercury	P	3E-10 kg
Methane, biogenic		0.00000004 kg
m-Xylene		0.00000012 kg
Nickel	P	0.000000006 kg
Nitrogen oxides		0.00007 kg
NM VOC, non-methane volatile organic compounds, unspe		0.0000023 kg
PAH, polycyclic aromatic hydrocarbons	P	1.11E-08 kg
Particulates, < 2.5 um	P	0.000026 kg
Phenol, pentachloro-		8.1E-12 kg
Phosphorus		0.0000003 kg
Potassium	P	0.0000234 kg
Sodium	P	0.0000013 kg
Sulfur dioxide	P	0.0000025 kg
Toluene		0.0000003 kg
Zinc	P	0.0000003 kg

Table A6. Case study 5: Life cycle inventory in ecoinvent for: Pellets, mixed, burned in furnace 15kW.

Table A7 lists all emissions to air for the Light fuel oil burning process used in case study 5. Substances labelled with “P” are assumed to be particles.

Heat, waste		1.0716 MJ
Acetaldehyde		2.05E-08 kg
Acetone		5E-08 kg
Acrolein		1.15E-08 kg
Hydrocarbons, aliphatic, alkanes, unspeci	P	2.5E-07 kg
Hydrocarbons, aliphatic, unsaturated	P	2E-08 kg
Hydrocarbons, aromatic	P	2E-08 kg
Benzaldehyde		6E-09 kg
Benzene		2E-08 kg
Butane		1.5E-07 kg
Methane, fossil		2E-07 kg
Carbon monoxide, fossil		0.000009 kg
Carbon dioxide, fossil		0.074 kg
Copper	P	4E-10 kg
Ethane		2E-08 kg
Ethene		5E-08 kg
Ethyne		1E-08 kg
Formaldehyde		6E-09 kg
Hydrogen fluoride		4.5E-09 kg
Mercury	P	5E-10 kg
Dinitrogen monoxide		7E-07 kg
Nitrogen oxides		2.75E-05 kg
PAH, polycyclic aromatic hydrocarbons	P	4.6E-10 kg
Particulates, < 2.5 um	P	5E-07 kg
Pentane		1E-07 kg
Propane		3E-08 kg
Propene		2E-08 kg
Propanal		6E-09 kg
Sulfur dioxide	P	4.57E-05 kg
Dioxin, 2,3,7,8 Tetrachlorodibenzo-p-	P	5.7E-17 kg
Toluene		1E-08 kg
Zinc	P	5E-10 kg

Table A7. Case study 5: Life cycle inventory in ecoinvent for: Light fuel oil, burned in boiler 10kW