Variations in the composition of house dust by particle size

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ABSTRACT
In this study, the distribution of heavy metals and other components in the various size fractions of house dust is investigated. A house dust sample collected from a vacuum cleaner was separated into size fractions by sieving and air classification. The analysis of the size fractions showed that the heavy metals and other components are not uniformly distributed in the various size fractions. The highest total carbon concentrations were found in the size fractions with a mass median diameter of 18–95 μm, while in the coarser size fractions and in the finest size fraction, the total carbon concentration was lower. In contrast, for many heavy metals and other metals (Al, Fe, Ca, S, Mn, Ti, Ba, Sr, As, Co, and V), the maximum concentrations were found in the finest size fraction. With increasing size of the dust fractions, the concentrations decreased. For several of these components, the dependence of the concentration on the particle size can be approximately assessed well using a power function. The distribution of Zn, Cu, Mg and Na was different. While the concentration of Na and Mg was higher in the coarser size fractions, no distinct trend was found for the concentrations of Cu and Zn.

Introduction
In industrialized countries, people spend approximately 88% of the time indoors in their homes and offices. Therefore, indoor contaminants like house dust can be significant in the intake of pollutants by humans. House dust is a heterogeneous mixture of organic and inorganic particles. The proportion of inorganic and organic matter in house dust may vary widely. A variation of the loss on ignition of house dust between 5 and 95% has been reported. Transport of particulate matter suspended in the atmosphere and soil adhering to footwear are the two main migration pathways of inorganic contaminants to indoors. The activities carried out within the house, especially heating, may be an additional important source of contaminants.

Over the past decades, there has been increasing concern about exposure of people to indoor contaminants, especially because of the amount of time people spend indoors. Heavy metals exist to a certain degree in the natural environment as trace elements in rocks and soils. However, they are released to the environment also from anthropogenic sources. They may originate from various sources, including traffic emissions and industrial sources. In relation to their crustal abundances, heavy metals show considerable enrichment in indoor environments. Values for the concentration of various heavy metals and other components in house dust have been reported in various studies. [6–13] Table 1 gives an overview of data reported in these studies.

Dust particles vary in shape and size. The particle size ranges from >2 mm to <63 μm with approximately one third of the dust being smaller than 500 μm. In previously published studies, the dust with a particle size of <500 μm or fractions of this dust were analysed. [6–13] The finer fractions of the house dust are small enough for uptake via inhalation or ingestion. [2]

The size dependence of the heavy metal concentrations is of special interest because the specific surface area of particles is indirectly proportional to the particle size. As mass transfer from a solid to a liquid takes place on the surface of the solid, the rate of mass transfer is higher in cases with finer particles, thus making hazardous components contained in these finer particles more available. Furthermore, the fraction of the very small particles in house dust (for example PM 2.5) is especially important with respect to inhalation after re-suspension. In few studies, the collected house dust was separated into size fractions before analysis. [14–16] Each of the three studies reports concentration data for three size fractions. Separation of the dust into the size fractions was performed by sieving. Thus, the separation of the fine dust was limited, and the finest size fraction produced was 0 μm to approximately 35 μm. The coarsest fraction was quite different in each study (48–63, 80–150 and 63–125 μm). The results show somewhat higher concentrations of Ni and Cr in the finest size fraction, while the concentrations of Al and Fe were higher in the coarsest fraction (Table 2). The results for Cu, Pb and Zn varied widely.

Different methods for sampling house dust are described in existing literature. The most common method used is vacuum sampling. [2,17]

The aim of this study was to investigate the distribution of heavy metals and other components in the various size...
Table 1. Average composition of house dust (in mg per kg of dry dust).

| Element | This study | Fergusson et al. [7] | Seifert et al. [12] | Mukerjee et al. [6] | Rasmussen et al. [9] | Chattopadhyay et al. [8] | Turner and Simmonds [11] | Karamelo [10] | Kurt-Karakus [13] |
|---------|------------|----------------------|---------------------|---------------------|----------------------|-------------------------|-------------------------|-------------|-----------------|
|         | Location of the study | Wels | Christchurch | Germany | USA | Ottawa | Sydney | UK | Elbasan | Istanbul |
|         | Year of the study | 2016 | 1996 | 1991–1992 | 1993 | 1993 | 1999 | 2005 | 2011 | 2012 |
|         | Phase-out of use of leaded gasoline | 1993 [18] | 1996 [19] | 1996 [18] | 1996 [18] | 1993 [18] | 2002 [10] | 2000 [20] | 2003 [21] | 2005 [22] |
|         | Number of house dust samples | 1 | 11 | 3894 | 9 | 48 | 82 | 32 | 6 | 31 |
|         | Size fraction analyzed | <500 μm | No data available | No data available | <53 μm | 100–250 μm | <100 μm | <63 μm | <500 μm | <149 μm |
| Al      | 3,703 | 23,900 | — | 30,850 | 25,948 | — | 8,740 | — | — |
| Ba      | 66 | — | — | 970 | 492 | — | — | — | — |
| Ca      | 66,700 | 14,900 | 5,170 | — | 48,760 | — | — | — | — |
| Co      | 9 | 8.6 | — | — | 8.9 | — | — | — | — |
| Cr      | 31 | 103 | 79.9 | 73.6 | 86.7 | 83.6 | — | 91.8 | 55 |
| Cu      | 190 | 230 | 118 | 126 | 206 | 147 | 339 | 195 | 156 |
| Fe      | 5,167 | 10,200 | 4,990 | 20,200 | 14,135 | 5,850 | 9,450 | 13,400 | — |
| K       | 2,460 | 12,600 | 3,880 | 13,170 | 10,305 | — | — | — | — |
| Mg      | 7,050 | 5,931 | 2,630 | — | 9,826 | — | — | — | — |
| Mn      | 234 | 207 | 144 | 322 | 269 | 76 | 578 | — | 136 |
| Na      | 7,610 | 11,800 | 5,610 | — | 23,224 | — | — | — | — |
| Ni      | 49 | — | — | — | 62.9 | 27.2 | 56.5 | 92.1 | 263 |
| Pb      | 29 | 734 | 56.9 | 240 | 405.6 | 389 | 181 | 73 | 28 |
| S       | 10,770 | — | — | 6,350 | — | — | — | — | — |
| Sr      | 57 | — | 44.6 | 436 | 255 | — | — | — | — |
| Ti      | 102 | 2,041 | — | 3,300 | — | — | — | — | — |
| V       | 12 | 30.4 | — | — | 24.8 | — | — | — | — |
| Zn      | 395 | 845 | 635 | 410 | 716.9 | 657 | 666 | 512 | 832 |
### Table 2. Average composition of size fractions of house dust.

| Location of the study | This study | Lisiewicz et al.\[^{14}\] | Hassan\[^{15}\] | Rasmussen et al.\[^{16}\] |
|-----------------------|------------|--------------------------|----------------|-------------------------|
| Year of the study     | 2016       | 1999                     | 2009           | 2001/2002               |
| Phase out of use of leaded gasoline | 1993\[^{18}\] | 2003\[^{22}\] | 1999\[^{23}\] | 1993\[^{18}\] |
| Number of house dust samples | 1          | 26                       | 4              | 63                      |
| Size fraction analyzed in g per kg of dry dust | <32 μm | 32–63 μm | 63–125 μm | <38 μm | 38–48 μm | 48–63 μm | <36 μm | 36–56 μm | 80–150 μm |
| AI                    | 12.3       | 7.59                     | 4.81           | 2.98                    | 2.18                   | 1.99                   | 207 | — | — | — | — | — | — | — | — |
| Ca                    | 168        | 106                      | 61.5           | 61.6                    | 56.1                    | 54.1                    | 75.1 | 54.4 | — | — | — | — | — | — | — | — |
| Fe                    | 14.6       | 11.0                     | 5.16           | 4.32                    | 4.67                    | 3.84                    | 4.12 | 4.13 | — | — | — | — | — | — | — | — |
| K                     | 3.02       | 4.09                     | 3.73           | 1.97                    | 1.37                    | 1.44                    | 1.50 | 1.59 | — | — | — | — | — | — | — | — |
| Mg                    | 3.75       | 6.05                     | 7.54           | 9.73                    | 3.62                    | 5.19                    | 4.57 | 7.47 | — | — | — | — | — | — | — | — |
| Na                    | 4.25       | 6.98                     | 8.36           | 5.49                    | 7.60                    | 7.75                    | 114  | 7.28 | — | — | — | — | — | — | — | — |
| S                     | 49.0       | 19.3                     | 12.8           | 9.44                    | 7.39                    | 5.13                    | 8.66 | 4.66 | — | — | — | — | — | — | — | — |
| TC                    | 186        | 289                      | 320            | 248                     | 152                     | 156                     | 165  | 164 | — | — | — | — | — | — | — | — |
| Size fraction analyzed in mg per kg of dry dust | <32 μm | 32–63 μm | 63–125 μm | <38 μm | 38–48 μm | 48–63 μm | <36 μm | 36–56 μm | 80–150 μm |
| AI                    | 278        | 97.8                     | 69.2           | 65.9                    | 51.9                    | 43.2                    | 496  | 43.3 | — | — | — | — | — | — | — | — |
| Ca                    | 41.6       | 15.7                     | 8.2            | 6.3                     | 8.8                     | 7.5                     | 7.5  | 66  | — | — | — | — | — | — | — | — |
| Cr                    | —          | —                        | —              | —                       | —                       | —                       | 106  | 102 | 93 | 98 | 78 | 68 | — | — | — | — |
| Cu                    | 151        | 310                      | 226            | 218                     | 148                     | 64.0                    | 838  | 233 | 186 | 136 | 141 | 134 | 182 | 217 | 138 | 135 | 170 |
| Mn                    | 413        | 359                      | 272            | 240                     | 162                     | 147                     | 175  | 197 | — | — | — | — | — | — | — | — | — |
| Ni                    | —          | —                        | —              | —                       | —                       | —                       | 106  | 102 | 93 | 98 | 78 | 68 | — | — | — | — |
| Pb                    | 106        | 55.2                     | 36.2           | 23.6                    | 36.2                    | 10.1                    | 104  | 22.2 | 209 | 169 | 144 | 217 | 214 | 209 | — | — | — |
| Sr                    | 265        | 85.3                     | 52.7           | 50.1                    | 49.5                    | 44.3                    | 50.5 | 52.8 | — | — | — | — | — | — | — | — | — |
| Ti                    | 350        | 167                      | 124            | 106                     | 75.3                    | 57.8                    | 47.6 | 53.2 | — | — | — | — | — | — | — | — | — |
| V                     | 65.7       | 23.0                     | 11.8           | 7.7                     | 10.3                    | 8.6                     | 9.6  | 8.1  | — | — | — | — | — | — | — | — | — |
| Zn                    | 362        | 553                      | 505            | 484                     | 254                     | 205                     | 123  | 359 | 1,200 | 1,230 | 1,430 | 173 | 120 | 102 | 830 | 892 | 729 |
fractions of house dust. For separation of the house dust into size fractions, beside conventional sieving, air classification was applied to be able to perform separation of finer size fractions as 35 μm, which was the limit in previous studies.[14–16]

**Materials and methods**

**Sample collection**

The study was performed in the Austrian town of Wels, which is a medium-sized city with approximately 60,000 inhabitants. There is no emission-intensive large-scale industry in the town or nearby. The house dust samples were collected from the filter of the central vacuum cleaning system in a 280 m² floor area residential house in the suburbs of Wels. The pipe network of the central vacuum cleaner system is made of plastic. The vacuum cleaner is used regularly for cleaning the wooden floors, carpets and the upholstered furniture. The entire dust content of the dust filter of the vacuum cleaner, accumulated over a period of approximately three months (Winter and Spring 2016), was taken for analysis. In the laboratory, the dust was dried at 105°C for 12 h. The total mass of the house dust sample was 899 g (dry matter). The moisture content of the dust was 6.7%. This relatively high value might be explained by the fact that the filter of the central vacuum cleaning system is installed in a non-heated room in the basement of the house. Thus, the temperature of the evacuated air decreases, which increases the relative humidity of the air and consequently the adsorption of moisture on the dust.

On the one hand, the significance of the study is somewhat limited because only one dust sample has been investigated. On the other hand, the investigated sample is an average for the three month dust collection period of the vacuum cleaner.

**Particle size fractionation**

In a first sample preparation stage, the dust was sieved with a 2.0 mm sieve. To separate the agglomerates of fibres and the dust, some rubber balls with a diameter of approximately 25 mm were used in the sieving procedure. The total mass of dust passing through the 2 mm sieve was 413 g (dry matter). The residue on the 2 mm sieve consisted mainly of fibre agglomerates, coarse fraction sanding material and small pieces of plastics, for example parts of children’s toys. However, after the sieving, there was still some fine dust within the fibre agglomerates. Therefore, the mass fraction of the various size fractions is only an indicator. From visual inspection, a maximum loss of up to 10% of finer dust with the discharged coarse fraction >2 mm might be possible. In a second sample preparation stage, the material that passed through the 2.0 mm sieve was sieved using a sieve stack consisting of the following sieves: 500, 400, 315, 250 and 200 μm. The selection of a maximum sieve size of 500 μm was based on the maximum dust size used in previous studies on house dust.[6–13] The total mass of dust passing the 500 μm sieve was 328 g (dry matter). While the residues from the four finer sieves were analysed, the residue from the 500 μm sieve was discarded. In a further sample preparation step, the material that passed the 200 μm sieve was air classified into four size fractions. Thus, in total, eight size fractions resulted from the house dust smaller than 500 μm.

For sieving, a laboratory sieve shaker ANALYSETTE 3 PRO from Fritsch (Idar-Oberstein, Germany) was used, and for air classification, a laboratory classifier 100 MZR from Hosokawa Alpine (Augsburg, Germany) was used. A detailed description of the sequential classification procedure can be found elsewhere.[24]

Because of some erosion of material on the classifier wheel, the recovery ratio was significantly greater than 1 for the heavy metals contained in the wheel material (Cr, Ni and Mo). Therefore, these components could not be investigated in the air-classified size fractions.[25] The particle size distribution of the size fractions was measured using a HELIOS/RODOS laser diffraction instrument from Sympatec (Clausthal-Zellerfeld, Germany) with dry sample dispersion. The instrument was checked with a Symptec SiC-P600/06 standard with a target value for the mass median diameter of 25.59 μm. The measured value for the mass median diameter was 25.62 μm. Microscopic images of particles of the finest size fraction were taken with a scanning electron microscope TESCAN, type MIRA3 (Brno, Czech Republic).

**Chemical analysis**

All chemical analyses were measured in duplicate. The total carbon (TC) content of the dust was determined using a LiquiTOC system with a solid material extension from Elementar Analysysysteme (Langenselbold, Germany). The organic and inorganic carbon is transformed by combustion with air into CO₂, which is subsequently analysed. The system was calibrated using an Elementar Analysysysteme soil standard with 4.1% TOC/TC.

For the analysis of metals and S, the dust samples were dissolved by aqua regia digestion. The concentrations of the heavy metals and various other metals were measured by inductively coupled plasma optical emission spectroscopy (Ultima 2 from Horiba Jobin Yvon, Bensheim, Germany), while the concentration of alkaline and earth alkaline metals (Na, K, Mg, Ca) and that of S were measured by ion chromatography using a ICS-1000 system from Dionex (Sunnyvale, CA, USA). Details for the analytical method can be found elsewhere.[26]

**Results and discussion**

**Characterization of the house dust sample**

Table 1 shows the composition of the investigated house dust in comparison with the results obtained in various studies. In all studies, the dust samples were collected from the dust bags of vacuum cleaners. The comparison of the results of the different studies is somewhat restricted because of differences in the sample pre-treatment applied in these studies, resulting in different size fractions being analysed. In the study of Rasmussen et al.[9] the collected house dust samples were sieved with a 250 and a 100 μm screen. In the subsequent chemical analysis, the size fraction of the dust between 100 and 250 μm was analyzed. In contrast, Mukerjee et al.[6] Turner and Simmonds,[11] Chatopadhyay et al.,[8] Kurt-Karakus [13] and Karamelo[10] analyzed the passage of a 53, 63, 100, 149 and 500 μm screen, respectively. In Fergusson et al.[7] and Seifert et al.,[12] there is
no information available on the particle size of the investigated dust.

The concentrations of most heavy metals are in the same range. However, major differences are evident for the concentrations of Pb. These differences might be explained by the phase-out and ban of Pb additives to gasoline. All studies with an average Pb concentration higher than 100 mg/kg were conducted before the phase-out of Pb in fuel in the respective country. As the Pb additives to fuel have been a major source of Pb in the environment, the measured Pb concentrations are lower in the studies conducted after the phase-out of Pb in fuel. However, Seifert et al. reported a rather low Pb concentration in their study conducted before the final ban of Pb in fuel in Germany. A possible explanation could be the gradual phase-out in Germany, as the Pb concentration in fuel had already been substantially reduced at the time the study was performed.

As a result of the change in the Pb emissions, the relative standard deviation for the reported Pb concentrations is quite high (91%). In contrast, the relative standard deviation for the other metals investigated in the majority of the studies (7 or 8 studies) is significantly lower: Cr 18%; Cu 38%; Zn 22%; Fe 47% and Mn 68%. Quite high values for the relative standard deviation were also found for Ni and Ca. However, data for these elements were reported in only a few studies.

In the investigated house dust, the concentrations of Ca, Mg, Co and S were higher compared to the average of the reported concentrations, while the concentration of Cu was equal to the average. For all other components, the measured concentrations were below the average of the reported concentrations. The biggest deviation of the measured concentration in this study from the reported average concentration was found for Ca. This might be explained by the high water hardness in the area of the study.

The TC content of the house dust sample was 23.1%. This compares well with the 33% result of another study where a concentration of organic matter in floor dust was collected in offices. Considering a typical mass fraction of carbon in organic matter, the TC content is quite similar. In another study, the average total organic carbon (TOC) content of 63 house dust samples was 26.8 ± 5.6% and the total inorganic carbon (TIC) was 1.44 ± 0.96%. A similar result was found for the average of 32 house dust samples in the United Kingdom with a TC content of 28.5 ± 8.1%.

Characterization of the size fractions of the house dust sample

The mass fraction and the mass median diameter for the five dust size fractions produced by sieving are summarized in Table 3. The data measured for the four size fractions produced by air classification of the finest sieving fraction are also shown in the table. The particle size distributions of the four coarser size fractions produced by sieving and the four size fractions produced by air classification are shown in Figures 1a and b, respectively. Microscopic images of dust particles from the finest size fraction are shown in Figure 2.

The concentrations in the eight size fractions are summarized in Table 2. Generally, the TC content was higher in the smaller size fractions with the exception of the smallest fraction. The maximum of 32% was found in the size fraction with

| Size fraction | Mass fraction (%) | Mass median diameter in μm |
|---------------|------------------|-----------------------------|
| 1             | 8.5              | 430                         |
| 2             | 11.9             | 340                         |
| 3             | 10.7             | 270                         |
| 4             | 6.3              | 220                         |
| 5-8           | 62.6             | 55                          |

| Size fraction | Mass fraction (%) | Mass median diameter in μm |
|---------------|------------------|-----------------------------|
| 5             | 25.3             | 95                          |
| 6             | 28.1             | 37                          |
| 7             | 7.3              | 18                          |
| 8             | 1.9              | 1.4                         |

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a mass median diameter of 37 \( \mu \text{m} \). In the coarse fractions with a mass median diameter of \( >200 \ \mu \text{m} \), the TC content was approximately 15\%, and the lowest TC content was 11.5\%. In the finest fraction, the TC content was nearly in the same range as in the coarse fractions \( >200 \ \mu \text{m} \).

For several components, a distinct dependence of the content on the particle size was found. For Ca, S, Fe, Al, Mn, Ti, Ba, Sr, Pb and to a lesser extent for K, the content decreased with increasing particle size, while for Na and Mg, the concentration increased slightly with the particle size (Table 2). The concentration of Zn and Cu also varied but showed no distinct trend. When the dependence of the concentration on the particle size is approximated by a power function of the type:

\[
c_i = \hat{k}_i \left( \frac{d_{50}}{d_{50i}} \right)^{a_i},
\]

where \( c_i \) is the concentration of the component, \( d_{50} \) is the mass median diameter of the size fraction and \( k_i \) and \( a_i \) are component-specific constants; for several components, a good correlation was found. In case of a correlation coefficient of \( r^2 \geq 0.75 \), the approximation functions are shown in Figure 2. For Ca, S, Fe, Al, Mn, Ti, Ba, Sr, V, Co and Pb, the correlation coefficient was 0.80, 0.94, 0.87, 0.97, 0.84, 0.97, 0.96, 0.83, 0.88, 0.84 and 0.75, respectively. For the other components, the correlation was much less distinct. A high value of the exponent \( a_i \) indicates a strong dependence of the concentration on the particle size. The exponent was highest for S, Pb, V, Al, Ti, Ba and Co with values of 0.38, 0.36, 0.35, 0.34, 0.32 and 0.30, respectively, and lower for Sr (0.28), Fe (0.24), Ca (0.19) and Mn (0.18).

The concentration of Cu and Zn also varied with the particle size. However, no distinct trend was found for this dependence. The concentrations of both components were higher in the fine size fractions and lower in the coarse ones. However, in the coarsest size fraction the concentrations were nearly as high as in the fine size fractions. The courses of the concentrations as a function of the particles size were quite similar for both components suggesting the possibility of similar sources for Cu and Zn in the house dust.

For comparison with the data available in the literature on the composition of house dust size fractions summarized in Table 2, the fine fraction, the medium fraction and the coarse fraction correspond approximately with the size fractions 7, 6 and 5 of this study. For Al and Fe, the results from this study are quite the opposite of the results reported by Hassan.\[15\] The concentrations of both elements decrease with increasing particle size in this study, while in Hassan,\[15\] an increase of the concentration with the particle size was reported. For Co, a decreasing concentration was also found in this study, while in Hassan,\[15\] the concentration was independent of the dust particle size.

The findings for Cu and Zn in the three studies are divergent (Table 2). While Hassan \[15\] and Rasmussen et al.\[16\] reported higher concentrations of Cu in the coarser dust, Lisiewicz et al.\[14\] found an opposite behaviour, which was confirmed in this study. For Zn, the findings are even more divergent. While Lisiewicz et al.\[14\] reported higher concentrations in the coarser dust, Hassan \[15\] found an inverse behaviour, and in the study of Rasmussen et al.,\[16\] no distinct trend was observed. In this study, a slight decrease in the Zn concentration with increasing particle size was observed in the corresponding particle size range.

For Pb, the results of this study are similar to those in the study by Lisiewicz et al.\[14\] Considerably lower Pb...
concentrations were found in the coarser size fractions of the dust. In contrast, Hassan [15] reported a constant concentration of Pb.

For most elements, the maximum concentration was found in the finest size fraction 8, which is more or less equivalent in particle size to the size dust fraction PM 2.5. The enrichment of components can be quantified by the enrichment factor, which is calculated as the quotient of the concentration of a component in the finest fraction and its concentration in the whole house dust <500 µm. The highest enrichment factors were found for V (5.7), Co (4.8), S (4.6), Ba (4.2), Sr (4.6) and Pb (3.7). For Ti, Al, Fe, Ca and Mn, the respective enrichment factors of 3.4, 3.3, 2.8, 2.5 and 1.8 were somewhat lower. For K, Cu and Zn, the concentration in the finest fraction was nearly those of the whole house dust <500 µm, while for Na and Mg, the concentration in the finest fraction was only half that concentration.

Conclusions

Heavy metals, TC and mineral components are not uniformly distributed in the various size fractions of house dust. The TC content varied between 11.5 and 32%. The highest concentrations were found in the size fractions with a mass median diameter of 18–95 µm, while in the coarse size fractions and in the finest size fraction, the TC content was lower.

In contrast, for many metals like Al, Fe, Ca, Mn, Ti, Ba and Sr, for S and for the heavy metals Pb, Co and V, the maximum concentrations were found in the finest size fraction. With increasing particle size, their concentrations in the dust decreased. For several of these components, the dependence of the concentration on the particle size can be approximated well using a power function.

The distribution of the concentrations of Zn, Cu, Na and Mg was divergent. For Na and Mg, the concentrations increased with increasing particle size, while for Zn and Cu, no distinct trend could be found.

A considerable enrichment of some heavy metals in the finest fraction of house dust was observed with enrichment factors of up to 5.7 in relation to the total dust <500 µm. This enrichment is expected to increase the rates of uptake via inhalation or ingestion of these components because of the inverse relation between specific surface area and particle size. Thus, in future studies, a special focus should be laid on the very fine fractions of the house dust.

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