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Review Article

The Nature of the Chemical Bond in Linear Three-Body Systems: From I_3^- to Mixed Chalcogen/Halogen and Trichalcogen Moieties

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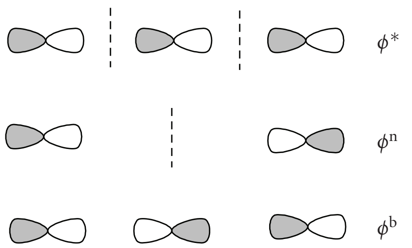
The 3 centre-4 electrons (3c-4e) and the donor/acceptor or charge-transfer models for the description of the chemical bond in linear three-body systems, such as I_3^- and related electron-rich (22 shell electrons) systems, are comparatively discussed on the grounds of structural data from a search of the Cambridge Structural Database (CSD). Both models account for a total bond order of 1 in these systems, and while the former fits better symmetric systems, the latter describes better strongly asymmetric situations. The 3c-4e MO scheme shows that any linear system formed by three aligned closed-shell species (24 shell electrons overall) has reason to exist provided that two electrons are removed from it to afford a 22 shell electrons three-body system: all combinations of three closed-shell halides and/or chalcogenides are considered here. A survey of the literature shows that most of these three-body systems exist. With some exceptions, their structural features vary continuously from the symmetric situation showing two equal bonds to very asymmetric situations in which one bond approaches to the value corresponding to a single bond and the second one to the sum of the van der Waals radii of the involved atoms. This indicates that the potential energy surface of these three-body systems is fairly flat, and that the chemical surrounding of the chalcogen/halogen atoms can play an important role in freezing different structural situations; this is well documented for the I_3^- anion. The existence of correlations between the two bond distances and more importantly the linearity observed for all these systems, independently on the degree of their asymmetry, support the state of hypervalency of the central atom.

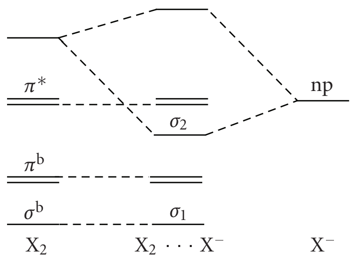
1. INTRODUCTION

The chemical bond in linear three-body systems, such as trihalides, has been the object of many papers appeared very recently in the literature [1–5]. Among them, the paper on trihalides and hydrogen dihalides published by Hoffmann et al. [2], the book edited by Akiba [1] "Chemistry of hypervalent compounds" appeared in 1999, and the chapter on hypervalent chalcogen compounds by Nakanishi [4] in "Handbook of chalcogen chemistry" edited by F. A. Devillanova, represent authoritative contributions to this topic. In particular, the paper by Hoffman analyzes, on the basis of theoretical calculations, the various contributions to the stabilization of trihalides, by comparing the Rundle-Pimentel [6, 7] model for electron-rich 3-centre 4-electron systems (Scheme 1) with that describing the interhalogenic bond as a donor/acceptor or charge-transfer interaction between a halide and a dihalogen molecule (Scheme 2).

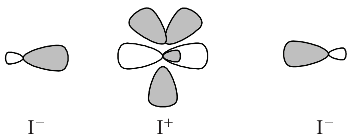
The commonly accepted 3 centre-4 electron bonding model considers the central halogen to be hypervalent. According to this model, a linear system formed for example by three aligned closed-shell I^- (24 shell electrons overall, I_3^{3-}) has no reason to exist since the three MOs generated by the combination of the three p_z orbitals, one from each interacting anion (Scheme 1), should be fully occupied by six electrons. However, the removal of two electrons from the antibonding MO causes an effective stabilization of the system and affords the well-known 22 shell electrons I_3^- anion (Scheme 1). The stability of I_3^- is determined by the occupancy of the lowest MO since the second filled MO is nonbonding in nature. The four electrons on the σ MOs plus the 6 electrons on the other three filled atomic orbitals equal a



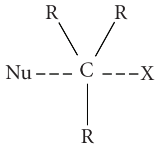
Scheme 1: Rundle-Pimentel model for electron-rich 3c-4e systems.



Scheme 2: First approximation MO diagram for the donor/acceptor interaction between X^- and X_2 fragments.



Scheme 3: Donation of two lone pairs from two I^- into two empty hybrid orbitals around I^+.



Scheme 4: Transition state of an SN_2 type reaction at an sp^3 carbon atom (Nu = nuclephilic group, X = leaving group).

total of 10 electrons on the central I atom; this accounts for its description as an hypervalent species. However, a more formal counting indicates that only 8 electrons can be assigned to the central iodine since the HOMO is a combination out of phase of the p_z orbitals of the external iodine atoms; consequently, no electron density coming from this MO can be assigned to the central iodine.

The alternative description of I_3^- according to a Lewis model, which considers an sp^2 I^+ cation having three lone pairs on the plane perpendicular to the bond direction and two hybrid orbitals able to linearly coordinate two I^- anions (Scheme 3), leads again to assign 10 electrons to the central iodine.

However, this model implies the combination of the d_z2 and p_z atomic orbitals of the central I^+ to afford two unfilled hybrid orbitals able to accommodate two lone pairs from the two I^- anions. According to this description, a simple notation, introduced by Arduengo et al. [5], identifies the central I atom as a 10-I-2 species, where "10" indicates the number of the electrons around the cental I, and "2" the number of the atoms or groups bonded to it. We must note that this formalism accounts very well for the hypervalent nature (expansion of the octet) of the central iodine, but does not account for the strength of the two bonds formed; in fact, according to such a model, each bond, arising from the localization of a pair of electrons from I^-, should have a bond order of 1.

The simplest way to prepare a triiodide or in general a trihalide species considers the reaction between an X^- anion with an X_2 molecule ($X^- + X_2 \rightarrow X_3^-$). In terms of chemical bond description, this corresponds to the commonly named donor/acceptor interaction (Scheme 2) since the bond is formed via a σ donation from one of the four filled atomic orbitals of X^- (np) towards the empty σ^* antibonding molecular orbital of X_2. As X^- formally approaches X_2, the three lone pairs of the approached X atom of the X_2 molecule are reoriented in order to be on the plane perpendicular to the bond direction when the symmetric three-body system X_3^- is formed. It corresponds to the rearrangement of a sp^3 carbon atom during a nucleophilic attack in an SN_2 type reaction (Scheme 4). The substantial difference between a trihalide and a penta-coordinated carbon resides in their different stabilities: while the penta-coordinated carbon represents a transition state, which finds its stabilization by removing one of the two apical groups so to allow the carbon to return to an sp^3 hybridization, X_3^- is a stable species. In such systems, the counting of the electrons around the central halogen and carbon atoms agrees very well with the notation by Arduengo et al. [5] (10-X-2 and 10-C-5, resp.). In fact, since the starting diatomic species (X_2 in the case of trihalides formation) obeys to the octet rule, every interaction with a donor (X^- in the case of trihalides formation) implies a transfer of electron density on X_2, thus formally justifying a number of electrons higher than 8 on the central atom of the resulting three-body system.

The simplified donor/acceptor first approximation MO diagram for the formation of a trihalide species (Scheme 2) becomes more complicate if the donor atomic orbital of X^- (np) is combined with both the σ^* and the σ^b MOs of X_2. The result is a second approximation MO diagram having three new energy levels for the adduct, coming from the combination of these three orbitals (Scheme 5).[1]

The difference between the first and second approximation MO diagrams (Schemes 2 and 5) resides in the nature of the first two MOs of the formed three-body system. In fact, the energy mixing in the second approximation diagram

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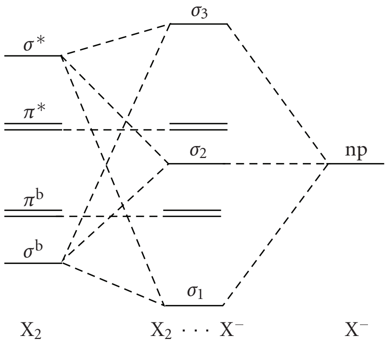
[1] However, in the construction of the simplified MO diagram for the donor/acceptor interaction (first approximation; Scheme 2), the combination of the donor orbital with only the σ^* MO of X_2, leaving unchanged the σ^b level, is justified fairly well by a good match of energy between the lone pair of X^- and the σ^* MO of X_2.



Scheme 5: Second-order approximation MO diagram for the donor/acceptor interaction between X^- and X_2 fragments (combination of np(X^-) with both σ^b and σ^* of X_2).

(Scheme 5) has the consequence of increasing the bonding nature of the lowest MO, moving the intermediate MO to higher energies towards a nonbonding nature. Now we can compare the 3c-4e bond model (Scheme 1) with the two MO diagrams for the donor/acceptor interaction between X^- and X_2 (Schemes 2 and 5) to describe the chemical bond in X_3^- anions. Since in all the three schemes, the highest MO is always an antibonding molecular orbital featuring a nodal plane between each couple of atoms, the differences between these models are mainly determined by the different nature of the lowest two molecular orbitals. According to the 3c-4e model, the stabilisation of electron-rich (22 shell electrons) three-body systems has to be ascribed only to the filling of the lowest MO, with a consequent bond order of 0.5 for each of the two bonds formed. According to the charge-transfer model, involving only the combination of a lone pair of X^- with the σ^* MO of X_2, the filling up of the lowest energy level corresponds to a bond order of 1 within the X_2 fragment, while the filling up of the intermediate level accounts for the bond formation between the two interacting fragments and for a lengthening of the X−X bond in X_2: a bond order of 0.5 for both bonds is reached in the symmetric situation $X-X-X^-$. In the second approximation charge-transfer MO diagram (Scheme 5), the lone pair of X^- combines with both σ^b and σ^* MOs of X_2, consequently the bonding nature of first molecular orbital of the resulting three-body system is increased, and that of the second energy level is decreased, thus making this MO diagram intermediate between the 3c-4e and the first approximation charge-transfer MO diagrams (Schemes 1, 2, and 5).

2. DISCUSSION

The two bonding models (3c-4e and charge-transfer models) can be successfully employed to describe the chemical bond in numerous linear three-body systems featuring 22 shell electrons, formed by three aligned main group elements. The 3c-4e model describes linear three-body systems (electron-rich linear systems) in terms of interacting aligned closed-shell fragments; the stabilization is reached by removing a couple of electrons in order to leave unfilled the highest MO (Scheme 1). Scheme 1 shows the combination of three p orbitals lying at the same level of energy; in a more general scheme with different starting closed-shell fragments, the combined p orbitals lie at different energy levels with the consequence that they will contribute differently to each molecular orbital in the resulting three-body system. In particular, if the combined p orbital of one of the two external atoms lies at an energy level quite different from that of the p orbitals of the other two atoms, its contribution to the bonding MO will be poor with a consequent unbalancing of the two bonds. This case is normally better described with the charge-transfer model, which corresponds to the interaction between a donor and a 2c-2e bond system.

Another aspect that we must consider is the total charge brought by the final three-body system; it will depend only on the charges of the starting aligned closed-shell species. A very simple example is represented by the formation of the XeF_2 molecule according to a 3c-4e model: the three-closed shell species to be considered are 2 F^- and Xe; by removing a couple of electrons the neutral XeF_2 molecule is generated. When three equal or different X^- (X^- = halide) are aligned, the resulting three-body system will be a trihalide monoanion.

The situation is much more complex for the formation of three-body systems from closed-shell species of 16th group elements, since the closed-shell species which can be combined can be both charged (E^{2-}, $R-E^-$) and neutral (R_2E and $R=E$, R = organic framework and E = chalcogen atom).[2] In general, the alignment of three identical chalcogen species can afford three-body systems featuring very different charges, (a)–(d) in Scheme 6.

In principle, any combination of E^{2-}, $R-E^-$, R_2E, and $R=E$ species is possible, thus strongly increasing the variety of obtainable three-body systems. In addition, the central chalcogen atom of the three-body systems (a)–(d) reported in Scheme 6 can be aligned in turn to one or two other couples of closed-shell chalcogen species to form, after removal of 1 or 2 couples of electrons, systems featuring two ((e)–(g) in Scheme 6) or three orthogonal 3c-4e fragments, respectively. In this way we can explain the great variety of structural archetypes which contain a hypervalent chalcogen atom. The number of possible combinations further increases if mixed S, Se, and Te systems are also taken into account (see below).

Analogously to asymmetric trihalides, many asymmetric trichalcogen and mixed dichalcogen/halogen and chalcogen/dihalogen systems can be successfully described using the same charge-transfer model as that used for asymmetric trihalides. According to this model three-body systems arise from the interaction between a donor species (halide or chalcogen) and an acceptor species (dihalogen,

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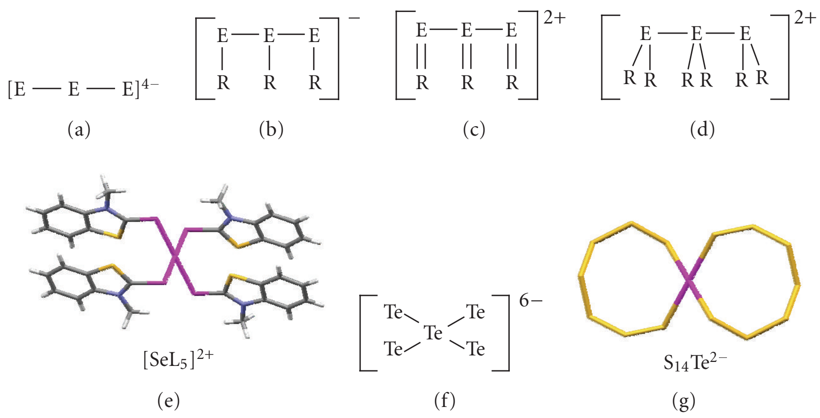
[2] In the case of 17th group elements, not only X^- but also $R'-X$ (R' represents a generic fragment featuring whichever element bonded to the halogen such as C−X, P−X, ...) closed-shell species could be considered in the formation of three-body systems; however, these cases are not considered here in order to limit the discussion.



SCHEME 6: Different three-body systems featuring aligned chalcogen atoms. In (e) L = N-methylbenzothiazole-2(3H)-selone.

dichalcogen, or chalcogen-halogen).[3] Therefore, a trichalcogen arrangement derives from the n(E)→ σ^*(E−E) interaction between one of the above-mentioned closed-shell chalcogen species acting as a donor and the empty σ^* MO of a dichalcogen molecule acting as an acceptor. In the case of mixed halogen/chalcogen systems, depending on the starting species, different topologies of three-body systems can be obtained, such as E−X−Y, X−E−Y, E−E−X, and E−X−E (E = chalcogen, X, Y = halogen), which correspond to the well-known charge-transfer adducts between chalcogen donors and dihalogens (E−X−Y), "T-shaped" adducts of chalcogen donors (X−E−Y), dichalcogen molecules interacting with halides (E−E−X), and halogen(+) linearly coordinated by two chalcogen donor molecules (E−X−E).

3. TRIHALIDES

The Cambridge Structural Database (CSD) has been searched for discrete trihalides fragments contained in deposited crystal structures; the results of the search are collected in Table 1.[4]

The triiodides are the most numerous and the scatter plot of the corresponding two I−I bond lengths is shown in Figure 1.

The literature related to triiodides has been omitted here and we refer to the paper by Svensson and Kloo [3]. Although several data are spread apart in the scatter plot,[5] the majority of them are concentrated in the region corresponding to symmetric or weakly asymmetric triiodides. It is important to point out that an analogous correlation is found for Br_3^- anions (see Figure 2) [44–109] while for other trihalides, such as ICl_2^- (Figure 3) [115–152] and IBr_2^- (Figure 4)

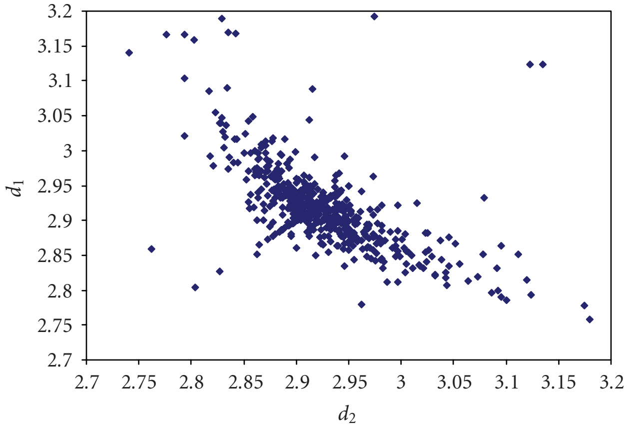


FIGURE 1: Scatter plot of d_1 versus d_2 for linear (angle > 165°) triiodides from a search of the CSD (608 structures containing 815 fragments). The mean bond lengthening is 9.7% with respect to the sum of the covalent radii.

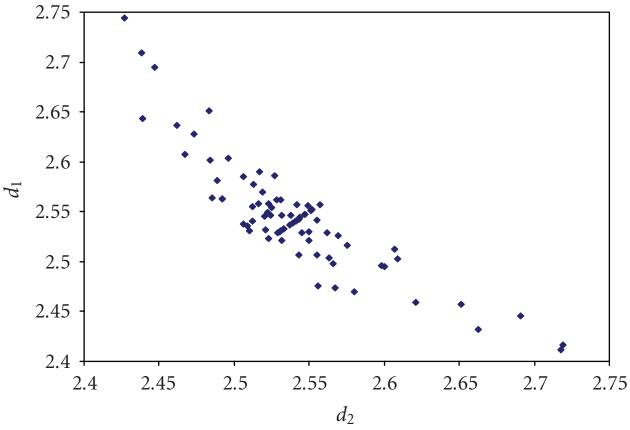


FIGURE 2: Scatter plot of d_1 versus d_2 for linear (angle > 165°) tribromides from a search of the CSD (71 structures containing 86 fragments). The mean bond lengthening is 11.3% with respect to the sum of the covalent radii.

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[3] As an exception, we will also consider the *pseudo*-halogens X−CN (X = halogen atom, see below in the last section).

[4] Only covalent bonded linear X−Y−Z fragments (angles > 165°) have been considered in subsequent analyses.

[5] A discussion on the factors responsible for the spreading of some data out of the correlation in Figure 1 is out of the aim of this work.

Table 1: Occurrence of linear isolated trihalide X−Y−Z fragments crystallographically characterized from a search of the Cambridge Structural Database (number of crystal structures in parentheses).

		Y−Z =					
		I−I	I−Br	I−Cl	Br−Br	Br−Cl	Cl−Cl
X =	I	809 (608)[a]	*	*	—	—	—
	Br	5[b]	56 (40)[c]	*	86 (71)[d]	—	—
	Cl	2[e]	4[f]	55 (46)[g]	—	1[h]	6[i]

[a] For the references of triiodides see [3]. [b] References [8–11]. [c] References [12–43]. [d] References [15, 44–109]. [e] References [10, 110, 111]. [f] References [112–114]. [g] References [32, 35, 43, 110, 111, 114–152]. [h] Reference [153]. [i] References [154–160]. *These fragments are already considered in the table.

Table 2: Structural features of all the less common X−Z−Y linear trihalides characterized by X-ray diffraction analysis.

Compound reference code	X	Z	Y	d(X−Z) (Å)	d(Z−Y) (Å)	∠X−Z−Y(°)*	References
CUPTIQ	Cl	Cl	Cl	2.182	2.394	177.7	[154, 155]
DEGLIK	Cl	Cl	Cl	2.248	2.338	177.5	[156]
PHASCL	Cl	Cl	Cl	2.227	2.306	177.4	[157]
UHUQAP	Cl	Cl	Cl	2.144	2.419	178.1	[158]
ZEHTIP	Cl	Cl	Cl	2.262	2.307	178.4	[160]
TEACBR	Cl	Br	Cl	2.379	2.401	176.8	[153]
DOBTUJ	Cl	I	Br	2.648	2.651	179.6	[112]
DOBTUJ04	Cl	I	Br	2.670	2.675	179.4	[113]
DOBTUJ07	Cl	I	Br	2.673	2.665	179.6	[114]
DOBTUJ08	Cl	I	Br	2.670	2.662	179.8	[114]
BEQXEA	I	I	Cl	2.737	3.040	172.1	[110]
LACPUB	I	I	Cl	2.765	2.739	179.3	[111]
EKIHEL	I	I	Br	2.890	2.906	178.7	[8]
EYOVAP	I	I	Br	2.857	2.950	179.3	[9]
LACQAI	I	I	Br	2.775	2.856	178.7	[10]
LACQUEM	I	I	Br	2.780	2.857	176.6	[10]
WOPGOX	I	I	Br	2.786	2.794	179.2	[11]

*The angle values are rounded off to the first decimal digit.

[12–43], the corresponding scatter plots show a much less evident correlation. The structurally characterized Cl$_3^-$ fragments [154–160] are less than Br$_3^-$ and I$_3^-$ ones, and no bond length correlation diagram is presented for them. Except for the case reported by Gorge et al. [159] in which the two terminal chlorine atoms have significant contacts with two nitrogen atoms, in the other six reported structures containing the Cl$_3^-$ anion the two bonds are differently elongated, being 2.144/2.419 Å the bond distances found in the more asymmetric case (see Table 2). The number of structurally characterized mixed trihalides featuring two different terminal halogens (I−I−Br$^-$ [8–11], I−I−Cl$^-$ [110, 111], Cl−I−Br$^-$ [112–114]) is very small, and a unique example of Cl−Br−Cl$^-$ is reported in the literature (Tables 1 and 2) [153]. Among the considered fragments, we wish to emphasize the structural changes occurring on changing one of the terminal halogen from Cl, to Br and to I. Consider, for example, the I−Cl bond length in different trihalides: d(I−Cl) increases on passing from the symmetric Cl−I−Cl$^-$ (mean value 2.53 Å) to Cl−I−Br$^-$ (mean value 2.663 Å, Table 2) and Cl−I−I$^-$ (mean value 2.889 Å, Table 2) indicating an increase in the ionic character of this bond when the other terminal halogen changes from Cl to Br and to I. However, in all cases, the I−Cl bond lengths are

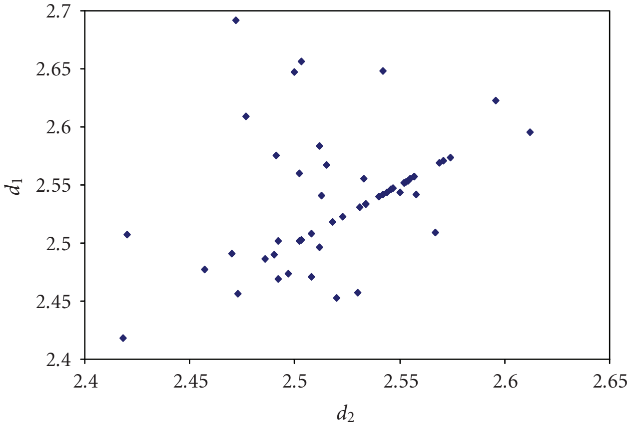


FIGURE 3: Scatter plot of d_1 versus d_2 for linear (angle > 165°) iododichlorides from a search of the CSD (46 structures containing 55 fragments). The mean bond lengthening is 9.2% with respect to the sum of the covalent radii.

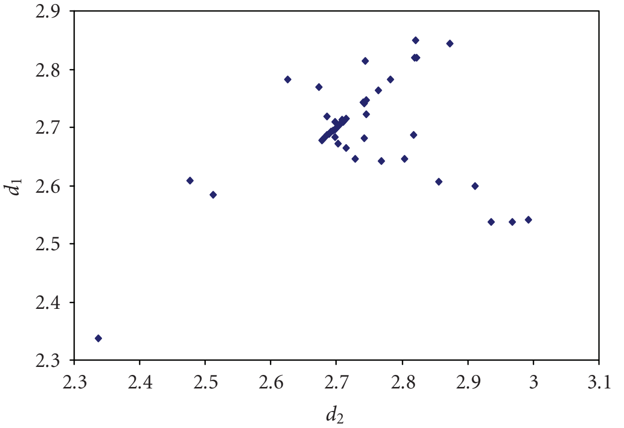


FIGURE 4: Scatter plot of d_1 versus d_2 for linear (angle > 165°) iododibromides from a search of the CSD (40 structures containing 56 fragments). The mean bond lengthening is 9.7% with respect to the sum of the covalent radii.

strongly elongated with respect to the sum of the covalent radii (2.39 Å) [161] but remain fairly shorter than the sum of the van der Waals radii (3.73 Å) [161]. The structural features of $I{-}I{-}Cl^-$ and $I{-}I{-}Br^-$ (Table 2) indicate that the bond distance of the central atom with the lighter halogen is always longer than the I−I distance, in accordance with a different ionic character of the two bonds. In terms of the 3c-4e model (Scheme 1), the *p* orbitals of the two terminal halogens do not contribute equally to the three molecular orbitals of the three-body systems $I{-}I{-}X^-$ ($X = Cl, Br, I$). In fact, the *p* orbital of the terminal atom featuring the better energy match with the *p* orbital of the central halogen will contribute more to the bonding MO; vice versa, the other halogen will mainly contribute to the nonbonding MO, thus carrying most of the negative charge. As a consequence, the bond orders of the two bonds diverge from the value of 0.5, one increasing towards the value of 1 (I−I), and the other decreasing towards the value of 0 (I−X). In terms of the charge-transfer model (Scheme 2), asymmetric trihalides of the type $X{-}Z\cdots Y^-$ derive from the donor/acceptor interaction between the halide (Y^-) and the acceptor species (X−Z); the strength of this interaction will depend on the reciprocal energy levels of the combining orbitals (*p* of the halide and σ^* MO of the dihalogen molecule).

However, in all trihalides, independently of the different polarization of the two bonds the sum of the bond lengths ($d_{X-Z} + d_{Z-Y}$) is always at least 9% longer than the sum of the covalent radii of the involved atoms, thus indicating a hypervalent state of the central halogen.

4. TRICHALCOGEN(IDE)S

Table 3 collects the occurrence of linear $E{-}E'{-}E''$ (E, E', E" = chalcogen atom) trichalcogen organic fragments found in structurally characterized compounds, as retrieved from a search of the Cambridge Structural Database (CSD) by imposing either the presence of two covalent bonds between the chalcogen atoms or the presence of one covalent bond and a nonbonding contact shorter than $\Sigma r_{VdW} - 0.3$ Å ($E\cdots E'{-}E''$ and $E{-}E'\cdots E''$ fragments). In both searches, the linearity of the fragment has been imposed ($\angle E{-}E'{-}E'' > 165°$).

As one can see, some combinations of trichalcogen systems have never been reported and some others have been found only in a limited number of structures (Table 4).

The scatter plots of $d(E{-}E')$ versus $d(E'{-}E'')$ for all trichalcogen fragments present in numerous crystal structures are shown in Figures 5–9.

Similar to what found for trihalides, linear trichalcogen systems can vary from symmetric to very asymmetric ones, but always feature strongly correlated $d(E{-}E')$ and $d(E'{-}E'')$ bond lengths. This indicates that also in linear trichalcogen $E{-}E'{-}E''$ organic fragments the potential energy hole should be fairly flat, being the chemical surrounding of the chalcogen atoms and the crystal packing effects able to freeze different structural situations. As mentioned above, in the case of 16th group elements, different closed-shell chalcogen species can interact to afford different types of linear trichalcogen systems (Scheme 6). However, since the analysis of all linear trichalcogen systems would go beyond the aim of this work, we will focus our attention only on some of them. When the closed-shell species are three E^{2-} anions, the corresponding three-body systems will be $E_3{}^{4-}$. Indeed, the linear $Te_3{}^{4-}$, together with the "T-shaped" $TeTe_3{}^{4-}$, and the square-planar $TeTe_4{}^{6-}$ anions are considered fundamental building units of numerous polytellurides [359]. The Te−Te bond distances in such tellurides show elongation of about 13% with respect to the sum of the covalent radii and are typical for 3c-4e bonds [359]. A symmetric $(Se_3)^{4-}$ ion was identified for the first time in the samarium/selenide cluster $[\{(C_5Me_5)Sm\}_6Se_{11}]$ [334], and considered a species isoelectronic to $I_3{}^-$. The Se−Se bond length in this system (2.749 Å) is much longer than the mean bond length in $(Se_2)^{2-}$ species (2.37 Å). This was justified by analogy with the couple $I_2/I_3{}^-$. Linear $[E{-}E\cdots E]^{4-}$ systems ($E = S, Se$) have been found in Mo and W clusters

Table 3: Occurrence of linear trichalcogen E−E′−E″, E · · · E′−E″, and E−E′ · · · E″ fragments crystallographically characterized from a search of the Cambridge Structural Database (number of crystal structures in parentheses).

		E′−E″ =					
		Te−Te	Te−Se	Te−S	Se−Se	Se−S	S−S
E =	S	3 (2)(a)	4 (3)(b)	207 (141)(c)	12 (7)(d)	16 (9)(e)	100 (64)(f)
	Se	2 (2)(g)	41 (24)(h)	∗	64 (43)(i)	∗	—
	Te	39 (27)(1)	∗	∗	—	—	—

(a)References [162, 163]. (b)Reference [164]. (c)References [164–250]. (d)References [251–256]. (e)References [251, 256–264].
(f)References [265–314]. (g)References [315, 316]. (h)References [162, 165, 174, 181, 183, 209, 216, 223, 228, 238, 317–323].
(i)References [201, 324–342]. (1)References [343–356]. ∗These fragments are already considered in the table.

Table 4: Structural features of less common E−E′−E″, E · · · E′−E″, or E−E′ · · · E″ trichalcogenides characterized by X-ray diffraction analysis.

Compound reference code	E	E′	E″	d(E−E′) (Å)	d(E′−E″) (Å)	∠E−E′−E″(°)(b)	References
BUWZUO	S	Se	S	2.266(a)	3.001(a)	172.2(a)	[259]
CEQKUE	S	Se	S	2.549	2.549	180.0	[260]
CUNWAJ	S	Se	S	2.534(a)	2.534(a)	180.0(a)	[261]
DUBKUG	S	Se	S	2.846(a)	2.295(a)	173.7(a)	[262]
FIKYUT	S	Se	S	2.467	2.371	170.0	[251]
KARZIM	S	Se	S	2.896(a)	2.282(a)	172.0(a)	[263]
SETIOP	S	Se	S	2.446	2.446	169.7	[264]
WAXMAJ	S	Se	S	3.302(a)	2.229(a)	169.2(a)	[256]
ZZZELOW01	S	Se	S	3.341	2.210	169.6	[257]
SOSNIX	S	Se	Se	3.002(a)	2.308(a)	167.3(a)	[253]
SOSNOD	S	Se	Se	2.977(a)	2.312(a)	167.3(a)	[253]
NPHSET	S	Se	Se	2.244	3.492	165.4	[254]
WADVOM	S	Se	Se	2.223	2.985	168.6	[255]
WAXMAJ	S	Se	Se	2.189	3.404	165.9	[256]
FIKYON	S	Se	Se	2.508	2.472	171.3	[251]
ZENJEH	S	Se	Se	2.498	2.466	173.6	[252]
FEZHIB	S	Te	Se	3.163	2.536	167.9	[162]
FEZHUN	S	Te	Se	2.592	2.872	175.3	[162]
FEZJEZ	S	Te	Se	3.002(a)	2.609(a)	173.4(a)	[164]
JOXYIE	S	Te	Te	3.508(a)	2.734(a)	170.4(a)	[162]
SISQUG	S	Te	Te	2.473	3.347	169.2	[163]
SEURBR(c)	Se	Se	Se	2.712	2.624	173.9	[337]
SEURSL(c)	Se	Se	Se	2.664	2.634	168.3	[339]
SECLUR(c)	Se	Se	Se	2.717	2.597	173.8	[337]
BAWFUA	Se	Te	Te	2.561	3.611	176.1	[315]
YOMRIB	Se	Te	Te	2.468	3.559	173.3	[316]
ZONWOO	O	Se	Se	2.427(d)	2.39	165.0	[357, 358]

(a)Mean values. (b)The angle values are rounded off to the first decimal digit.
(c)Triselenourea dications with different counterions. (d)d(O · · · Se).

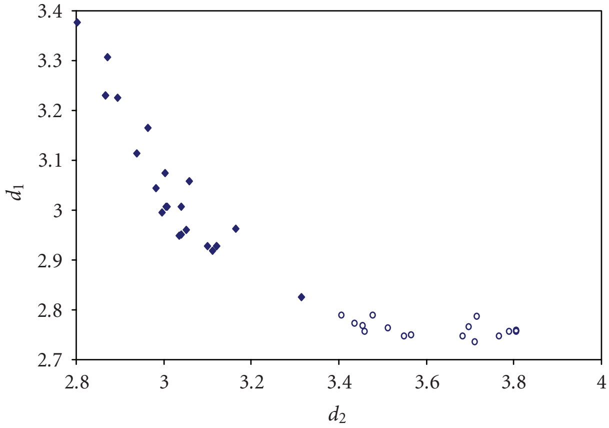


FIGURE 5: Scatter plot of d_1 versus d_2 for linear (angle > 165°) Te−Te−Te fragments from a search of the CSD. The symbol (◆) refers to the 23 Te−Te−Te fragments (19 structures) featuring bond distances ranging from Σr_{cov} to $(\Sigma r_{VdW} - 0.6)$; the symbol (∘) refers to the 16 Te···Te−Te fragments (12 structures) featuring Te···Te contact distances shorter than $(\Sigma r_{VdW} - 0.3)$. The mean bond lengthening within Te−Te−Te fragments is 11.5% (17.2% on Te···Te−Te fragments) with respect to the sum of the covalent radii.

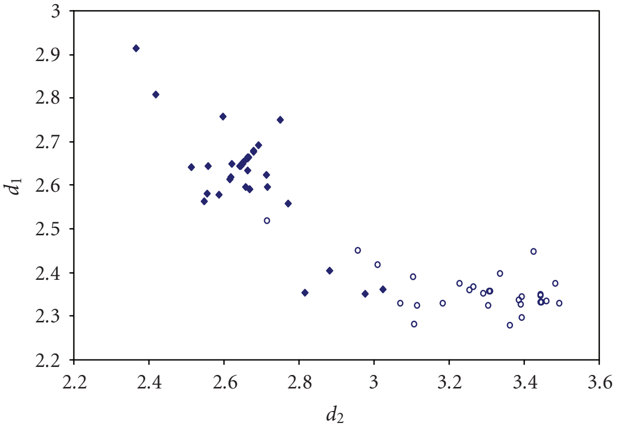


FIGURE 6: Scatter plot of d_1 versus d_2 for linear (angle > 165°) Se−Se−Se fragments from a search of the CSD. The symbol (◆) refers to the 35 Se−Se−Se fragments (22 structures) featuring bond distances ranging from Σr_{cov} to $(\Sigma r_{VdW} - 0.6)$; the symbol (∘) refers to the 29 Se···Se−Se fragments (21 structures) featuring Se···Se contact distances shorter than $(\Sigma r_{VdW} - 0.3)$. The mean bond lengthening within Se−Se−Se fragments is 13.9% (21.5% on Se···Se−Se fragments) with respect to the sum of the covalent radii.

[257, 335] containing two $[M_3(\mu_3\text{-}E)(\mu\text{-}E_2)_3(dtc)_3]^+$ cores (M = Mo, E = S, Se; M = W, E = Se) linked via an E^{2-} anion; these arrangements have been described as μ-${E_2}^{2-}$ dichalcogenides interacting with E^{2-} at significantly short distances. In the case of selenium clusters [335], the two Se−Se bonds are 2.355 Å and 2.816 Å for the Mo cluster and 2.38 Å and 2.93Å (mean values) for the W one, the short distances being only slightly elongated with respect to the Se−Se bond length in diselenides (2.34 Å). As found for strongly asymmetric trihalides, which are better described as an X^- anion interacting with an X_2 molecule ($X^- \cdots X_2$), the trichalcogen systems in which the two bonds assume very different bond orders should be better described as a chalcogen donor (in the present case E^{2-}) interacting with the σ^* antibonding molecular orbital of a dichalcogen species [n(E)→ σ^*(E−E), in the present case ${E_2}^{2-}$]. In other words, the interaction should occur between a chalcogen donor and a 2c-2e dichalcogen bond system. In general, depending on the starting chalcogen donor and dichalcogen acceptor species, these trichalcogen systems can carry a variable charge, from negative values as in the above cases, up to 2+ when the donor species is a neutral molecule and the acceptor a dichalcogen dication.

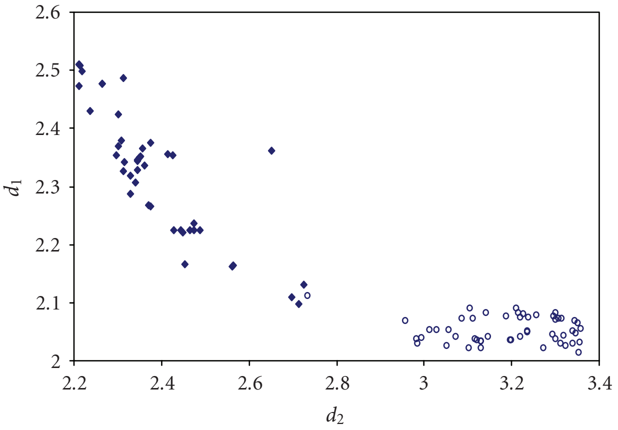


FIGURE 7: Scatter plot of d_1 versus d_2 for linear (angle > 165°) S−S−S fragments from a search of the CSD. The symbol (◆) refers to the 48 S−S−S fragments (40 structures) featuring bond distances ranging from Σr_{cov} to $(\Sigma r_{VdW} - 0.6)$; the symbol (∘) refers to the 52 S···S−S fragments (24 structures) featuring S···S contact distances shorter than $(\Sigma r_{VdW} - 0.3)$. The mean bond lengthening within S−S−S fragments is 15.5% (28.6% on S···S−S fragments) with respect to the sum of the covalent radii.

Several monoanionic structures of the type ${(R{-}E)_3}^-$, arising from three aligned $R{-}E^-$ anions, for example, ${(Ph{-}Te)_3}^-$ [343, 356], or $[(CN)Se{-}Se(Ph){-}Se(CN)]^-$ [209], have been reported. Numerous are the hypervalent chalcogen compounds deriving from a neutral species interacting with two negatively charged monochalcogenides such as the case of 2,5-bis(morpholino-N)-4a-phenyl-1,3a,6,6a-tetrahydro-1,6,6a-triselena-4aλ^4-phospha-3,4-diazapentalene [332], or from a chalcogenide(2−) interacting with neutral molecules to form 1 or more 3c-4e systems, as in the case of the tetrakis(N-methylbenzo thiazole-2(3H)-selone)selenium(2+) dication reported by us in which two orthogonal 3c-4e fragments are present [342]. Particularly interesting are the two organic compounds FIKYON [251] and ZENJEH [252] (Table 4) containing the linear Se−Se−S arrangement. In fact, in both cases, the Se−Se bond is shorter than the Se−S one due to the poor energy match between the orbital of the central Se atom and that of the peripheral S atom. When the closed-shell

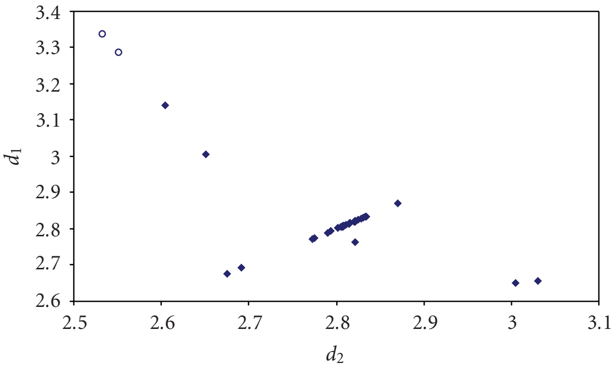


Figure 8: Scatter plot of d_1 versus d_2 for linear (angle > 165°) Se−Te−Se fragments from a search of the CSD. The symbol (◆) refers to the 39 Se−Te−Se fragments (22 structures) featuring bond distances ranging from Σr_{cov} to ($\Sigma r_{VdW} - 0.6$); the symbol (◦) refers to the 2 Se ⋯ Te−Se fragments (2 structures) featuring Se ⋯ Te contact distances shorter than ($\Sigma r_{VdW} - 0.3$). The mean bond lengthening within Se−Te−Se fragments is 11.5% (16.1% on Se ⋯ Te−Se fragments) with respect to the sum of the covalent radii.

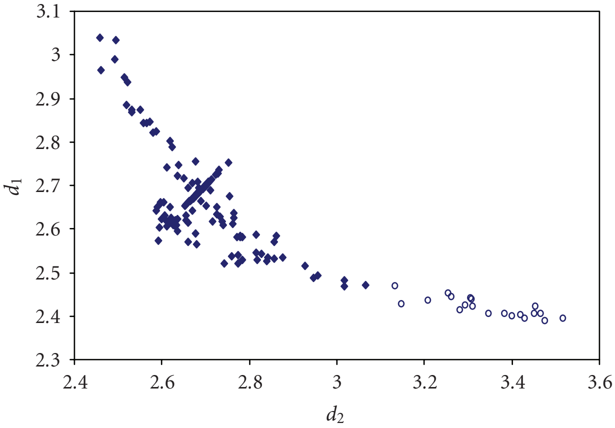


Figure 9: Scatter plot of d_1 versus d_2 for linear (angle > 165°) S−Te−S fragments from a search of the CSD. The symbol (◆) refers to the 187 S−Te−S fragments (127 structures) featuring bond distances ranging from Σr_{cov} to ($\Sigma r_{VdW} - 0.6$); the symbol (◦) refers to the 20 S ⋯ Te−S fragments (14 structures) featuring S ⋯ Te contact distances shorter than ($\Sigma r_{VdW} - 0.3$). The mean bond lengthening within S−Te−S fragments is 12.6% (21.0% on S ⋯ Te−S fragments) with respect to the sum of the covalent radii.

species are S-, Se-, or Te-containing neutral molecules, dicationic species will be generated having the central atom in a hypervalent state. Among these systems, those having three S−S−S aligned sulphur atoms [265–314] have been found only in the class of the pincer-type molecules, with the central sulphur able to bind or to move apart the terminal ones by oxidation/reduction processes. It is noteworthy to observe that most of the molecules belonging to the E−E′−E pincer-type arrangments and many other trichalcogen systems are fairly symmetric, even if examples of strongly asymmetric situations are also numerous.

Although our discussion is limited to the structural features of linear trichalcogen fragments (various combinations of S, Se, and Te), we have also included in Table 4 the only known example of an organic dichalcogen dication system having a strong contact with an oxygen atom [357, 358]. X-ray analysis of this dication confirmed the linear geometry of the O−Se−Se moiety (165°) and an Se−Se bond (2.39 Å) which, similarly to what found in the above-described Mo and W clusters [257, 335], is only slightly elongated with respect to an Se−Se bond in diselenides (2.34 Å). This compound represents a good example of a hypervalent selenium compound having the two bonds strongly unbalanced (bond orders very far from the value of 0.5 expected for a balanced 3c-4e bond system). For this reason it resembles many other similar systems, such as the adduct of N,N′-dimethylimidazoline-2-selone with the *pseudo*-halogen ICN recently reported by us (see below in the last section); in both cases, one of the bonds tends to be a single bond, while the other bond is very elongated and tends to assume a purely ionic character.

5. DICHALCOGEN-HALIDES

Two chalcogen and one halogen atoms as closed-shell species can be aligned in only two possible ways: the halogen in the terminal (E−E′−X) or in the central (E−X−E′) position. Both arrangements are known and they will be discussed separately.

5.1. E−E′−X *fragments*

Table 5 shows the number of linear E−E′−X fragments crystallographically characterized from a search of the Cambridge Structural Database, by imposing the linearity of the system (∠E−E′−X angle > 165°) and either the presence of two covalent E−E′ and E′−X bonds or the presence of one E−E′ covalent bond and one E′ ⋯ X nonbonding contact shorter than $\Sigma r_{VdW} - 0.3$ Å.

It is interesting to note that in the case of the S−S−X fragment (X = Cl, Br, I) the number of structures characterized by the presence of a linear S−S ⋯ X moiety is considerably higher than that featuring the S−S−X one (17 versus 4). Fragments having fairly covalent bonds have been found exclusively as part of some molybdenum clusters [390, 391, 399, 401, 445–447]. These clusters are very similar to those previously described in the discussion of trichalcogenides species, with the difference that the halide takes the place of the bridging E^{2-} anion. On the basis of their insolubility in water and their solubility in the common organic solvents, the authors concluded that the S−X bonds should be prevalently covalent in character. In fact, their structural features seem to be consistent with the presence of an $[S{-}S{-}X]^{3-}$ anion, deriving from the removal of a couple of electrons from the aligned S^{2-}, S^{2-}, and X^- closed-shell species. The sum of the S−S and S−X bond lengths in these fragments is about 23% longer than the sum of the covalent radii and about 31% shorter than the sum of the van der Waals radii, in agreement with a 3c-4e bond model. The scatter plots of d(S−S) versus

TABLE 5: Occurrence of linear E−E′−X and E−E′ · · · X fragments crystallographically characterized from a search of the Cambridge Structural Database (number of crystal structures in parentheses).(a)

		E−E′ =					
		Te−Te	Se−Te	S−Te	Se−Se	S−Se	S−S
X =	Cl	6 (3)(b)	5 (5)(c)	53 (39)(d)	11 (8)(e)	7 (6)(f)	49 (21)(g)
	Br	—	17 (11)(h)	35 (26)(i)	23 (11)(j)	4 (2)(k)	47 (17)(l)
	I	19 (7)(m)	4 (4)(n)	20 (19)(o)	5 (3)(p)	3 (1)(q)	32 (19)(r)

(a) Most of the structures have been found by imposing the presence of at least a contact between the E−E′ and X fragments (E−E′ · · · X) shorter than ($\Sigma r_{\mathrm{VdW}} - 0.6$). (b) References [360–362]. (c) References [317, 318, 363–365].
(d) References [173, 174, 188–190, 194, 200, 204, 205, 212, 213, 235, 244, 317, 363, 366–381]. (e) References [38, 360, 382–385].
(f) References [260, 386–389]. (g) References [390–410]. (h) References [317, 363, 364, 370, 411, 412].
(i) References [182, 196, 207, 212, 213, 232, 235, 317, 363, 365, 367, 372, 373, 377, 379, 413–418]. (j) References [38, 398, 419–425].
(k) Reference [386]. (l) References [398, 405, 426–433]. (m) References [434–438]. (n) References [214, 364, 439, 440].
(o) References [200, 213, 214, 235, 317, 372, 373, 378, 440–443]. (p) References [38, 444]. (q) Reference [383]. (r) References [399, 445–458].

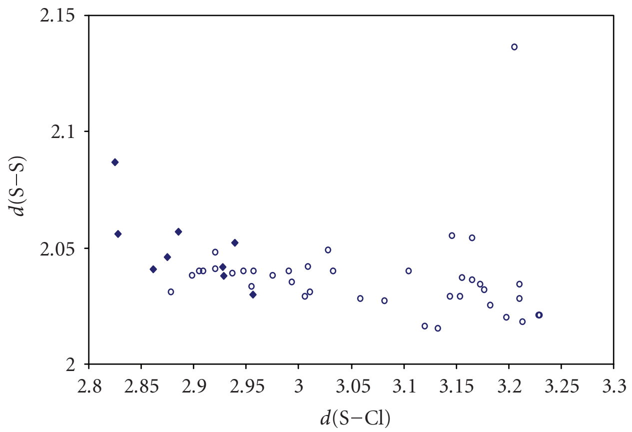


FIGURE 10: Scatter plot of $d(\mathrm{S{-}S})$ versus $d(\mathrm{S{-}Cl})$ for linear (angle > 165°) S−S−Cl fragments from a search of the CSD. The symbol (◆) refers to the 10 S−S−Cl fragments (4 structures) featuring bond distances ranging from Σr_{cov} to ($\Sigma r_{\mathrm{VdW}} - 0.6$); the symbol (○) refers to the 39 S−S · · · Cl fragments (17 structures) featuring S · · · Cl contact distances shorter than ($\Sigma r_{\mathrm{VdW}} - 0.3$). The mean bond lengthening within S−S−Cl fragments is 21.5% (26.1% on S−S · · · Cl fragments) with respect to the sum of the covalent radii.

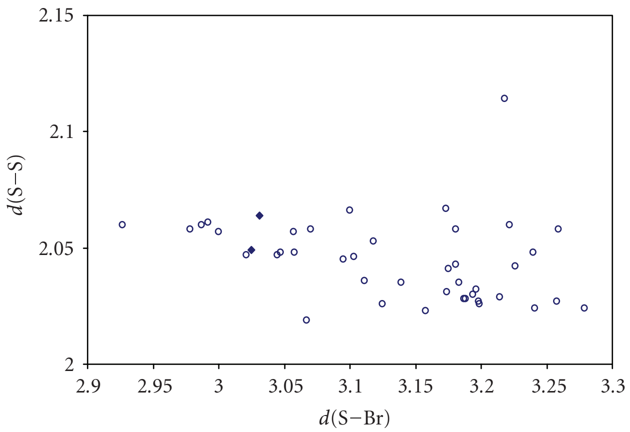


FIGURE 11: Scatter plot of $d(\mathrm{S{-}S})$ versus $d(\mathrm{S{-}Br})$ for linear (angle > 165°) S−S−Br fragments from a search of the CSD. The symbol (◆) refers to the 2 S−S−Br fragments (1 structure) featuring bond distances ranging from Σr_{cov} to ($\Sigma r_{\mathrm{VdW}} - 0.6$); the symbol (○) refers to the 45 S−S · · · Br fragments (16 structures) featuring S · · · Br contact distances shorter than ($\Sigma r_{\mathrm{VdW}} - 0.3$). The mean bond lengthening within S−S−Br fragments is 21.1% (23.7% on S−S · · · Br fragments) with respect to the sum of the covalent radii.

$d(\mathrm{S{-}X})$ for both S−S−X and S−S · · · X (X = Cl, Br, I) fragments are shown in Figures 10, 11, and 12.

It appears very clear from the figures that the points (◆) related to the S−S−X fragments are not enough to establish the existence of a correlation between the two bond distances, and those (○) related to the S−S · · · X fragments do not fit any correlation, since the S−S bond distances range in a very small interval of values around 2.05 Å (1.02 Å is the covalent radius of S), and the S · · · X contacts in a very wide interval of distances. These observations are more consistent with a description of these systems as deriving from donor/acceptor interactions between one of the p orbitals of X^- and the σ^* antibonding MO on the S−S system. In these systems the energy-match between the lone pair of X^- and the σ^* MO on $\mathrm{S_2}^{2-}$ is very poor. The weak interaction is reflected in the corresponding very low lengthening of the S−S bond.

Numerous crystal structures have been reported in the literature that feature linear S−Te−X (X = Cl, Br, I) systems (references are collected in Table 5). Contrary to what found on searching the CSD for S−S−X fragments, for these linear arrangements, almost all the fragments feature covalent S−Te and Te−X bonds (S−Te−X systems). Only one structure containing an S−Te · · · Cl [244] moiety has been found by searching for S−Te · · · X systems [$\Sigma r_{\mathrm{cov}} - 0.6 < d(\mathrm{T} \cdots \mathrm{X}) < (\Sigma r_{\mathrm{VdW}} - 0.3)$]. As shown in Figures 13, 14, and 15, for these three series of compounds the two bonds are strictly correlated in wide ranges of variability.

The sum of S−Te and Te−X bond distances within these three-body systems is 10.8%, 11.6%, and 7.3% longer than the sum of the covalent radii for X = Cl, Br, and I, respectively, in good agreement with the hypervalent nature of the central tellurium atom. The other mixed dichalcogen fragments bonded to a halide, characterized by X-ray diffraction, are very few and they are collected in Table 6.

Table 6: Structural features of the less common E−E′−X and E−E′ · · · X (E, E′ = S, Se, X = halogen) linear three-body systems and of some selected E−E′−X (E = S, Se, E′ = Te, X = halogen) fragments.

Compound reference code	E	E′	X	d(E−E′) (Å)	d(E′−X) (Å)	∠ E−E′−X(°)§	References
BOYXAO10	S	S	Cl	2.040*	2.915*	169.6*	[390]
FAVDUB	S	S	Cl	2.053*	2.863*	166.9*	[391]
KOJHOG	S	S	Cl	2.047*	2.933*	170.4	[399]
PIGWIL	S	S	Cl	2.087	2.825	165.1	[401]
				2.179	2.573	168.3	
KOJHUM	S	S	Br	2.056*	3.028*	171.5*	[399]
CIKHUZ10	S	S	I	2.057*	3.168*	171.2*	[445]
JAKWAT	S	S	I	2.057*	3.150*	173.1*	[446]
KOJJEY	S	S	I	2.066*	3.175*	172.6*	[399]
PEHHOZ	S	S	I	2.051	3.180	172.8	[447]
QADHOS	S	Se	I	2.218*	3.149*	168.3*	[383]
MURXOM	S	Se	Br	2.285*	3.007*	175.0*	[386]
MURYAZ	S	Se	Br	2.258*	3.094*	174.3*	[386]
MURXIG	S	Se	Cl	2.273*	2.920*	174.8*	[386]
MURXUS	S	Se	Cl	2.252	2.976	172.8	[386]
CEQKOY	S	Se	Cl	2.215	3.276	178.5	[260]
KAXWEL	S	Se	Cl	2.293	3.237	168.8	[387]
NEDBAZ	S	Se	Cl	2.136	3.212	171.9	[389]
TAVXET	Se	Se	Cl	2.440	2.778	172.1	[38]
TAVXIX	Se	Se	Br	2.424	2.830	166.7	[38]
PEBPUH	Se	Se	Br	2.403	3.036	174.2	[419]
WOHDUS	Se	Se	Br	2.529*	2.689*	174,4*	[420]
EZOYIB	Se	Te	I	2.906	2.889	177.7	[439]
FOBCEE	Se	Te	I	2.618	3.251	173.5	[364]
ISEUTE	Se	Te	I	2.679	3.095	177.3	[440]
ROMXEW	Se	Te	I	2.721	2.967	177.5	[214]
BSEUTE	Se	Te	Br	2.616	3.054	175.6	[370]
DEVHAN	Se	Te	Br	2.769	2.761	175.2	[363]
FOBBIH	Se	Te	Br	2.678	2.898	173.9	[317]
FOBBIH01	Se	Te	Br	2.673*	2.907*	173.7*	[317]
FOBCAA	Se	Te	Br	2.572*	3.096*	172.9*	[364]
FOBCAA01	Se	Te	Br	2.582	3.086	174.0	[364]
FOBCAB	Se	Te	Br	2.648	2.854	174.8	[364]
KIKPID	Se	Te	Br	2.496*	3.244*	168.6*	[411]
NAHWIC	Se	Te	Br	2.704*	2.810*	175.0*	[412]
NAWOI	Se	Te	Br	2.763	2.744	177.0	[412]
FOWMAF	Se	Te	Br	2.540	3.289	174.3	[318]
DEVGUG	Se	Te	Cl	2.783	2.600	174.7	[363]
FOBBED	Se	Te	Cl	2.678	2.752	172.8	[317]
FOBBUT	Se	Te	Cl	2.664	2.701	175.6	[364]
GANHIM	Se	Te	Cl	2.592	2.972	171.6	[365]
BETDAG	Te	Te	I	3.283	2.814	166.7	[434]
HOJJEV	Te	Te	I	3.163	2.831	176.1	[435]
HOJJEV01	Te	Te	I	3.158	2.817	175.6	[436]
HOSCAT	Te	Te	I	2.669*	3.369*	169.0*	[437]
HOSCEX	Te	Te	I	2.644*	3.329*	167.8*	[437]

§The angle values are rounded off to the first decimal. *Mean values.

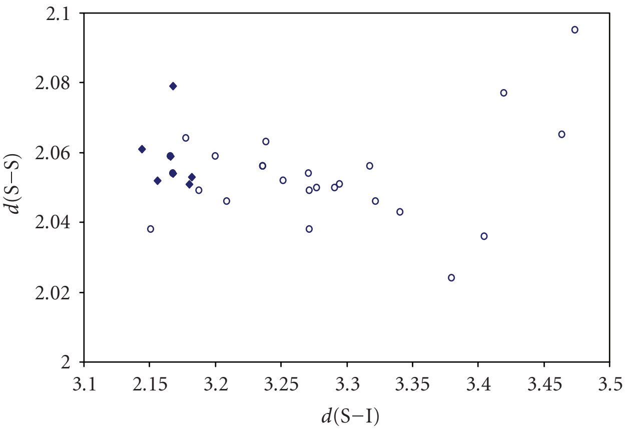


Figure 12: Scatter plot of d(S−S) versus d(S−I) for linear (angle > 165°) S−S−I fragments from a search of the CSD. The symbol (◆) refers to the 7 S−S−I fragments (4 structures) featuring bond distances ranging from Σr_{cov} to $(\Sigma r_{\text{VdW}} - 0.6)$; the symbol (∘) refers to the 25 S−S$\cdots$I fragments (11 structures) featuring S$\cdots$I contact distances shorter than $(\Sigma r_{\text{VdW}} - 0.3)$. The mean bond lengthening within S−S−I fragments is 19.2% (21.5% on S−S$\cdots$I fragments) with respect to the sum of the covalent radii.

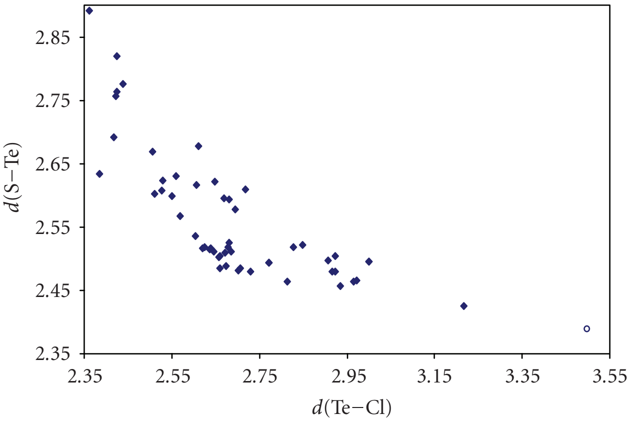


Figure 13: Scatter plot of d(S−Te) versus d(Te−Cl) for linear (angle > 165°) S−Te−Cl fragments from a search of the CSD. The symbol (◆) refers to the 52 S−Te−Cl fragments (38 structures) featuring bond distances ranging from Σr_{cov} to $(\Sigma r_{\text{VdW}} - 0.6)$; the symbol (∘) refers to the 1 S−Te$\cdots$Cl fragment (1 structure) featuring Te$\cdots$Cl contact distances shorter than $(\Sigma r_{\text{VdW}} - 0.3)$. The mean bond lengthening within S−Te−Cl fragments is 10.8% (24.5% on S−Te$\cdots$Cl fragment) with respect to the sum of the covalent radii.

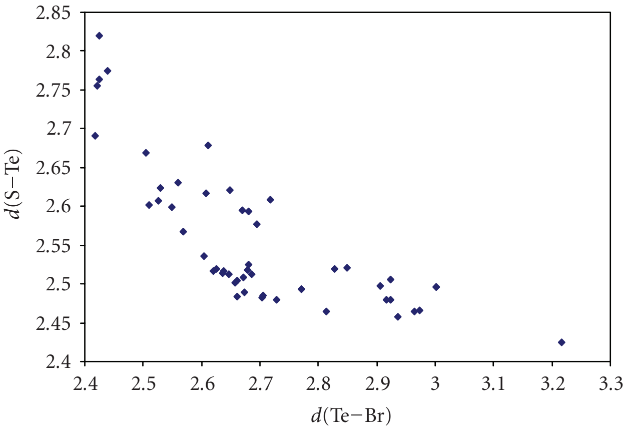


Figure 14: Scatter plot of d(S−Te) versus d(Te−Br) for the 52 linear (angle > 165°) S−Te−Br fragments (38 structures) featuring bond distances ranging from Σr_{cov} to $(\Sigma r_{\text{VdW}} - 0.6)$ from a search of the CSD. The mean bond lengthening within S−Te−Br fragments is 11.6% with respect to the sum of the covalent radii.

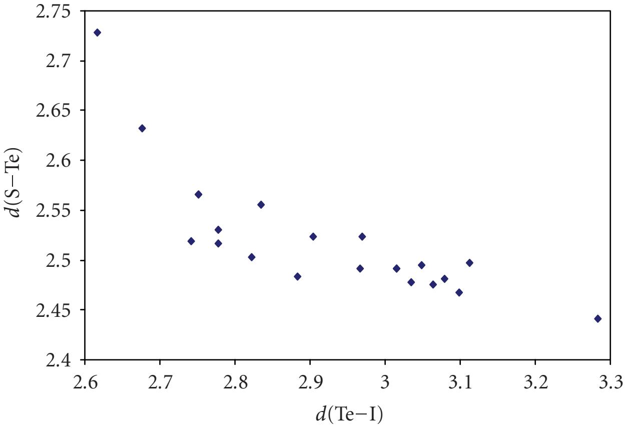
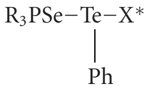


Figure 15: Scatter plot of d(S−Te) versus d(Te−I) for the 20 linear (angle > 165°) S−Te−Br fragments (19 structures) featuring bond distances ranging from Σr_{cov} to $(\Sigma r_{\text{VdW}} - 0.6)$ from a search of the CSD. The mean bond lengthening within S−Te−Br fragments is 7.3% with respect to the sum of the covalent radii.

Differently from S−S−X, the S−Se−X fragments have been found in some dimeric structures with bridging halides [383, 386]. Also in these cases the sum of the S−Se and Se−X bond lengths shows elongation (∼19%) with respect to the sum of the covalent radii, and shortening (∼30%) with respect to the sum of the van der Waals radii. A certain number of structures characterized by the linear Se−Te−X system have also been found. It is noteworthy that the S−Te and Se−Te bonds get shortened as the Te−X bond becomes more ionic (on changing X from I to Br and to Cl, see the examples reported in Table 7), their bond orders approaching the value of 1.

In the case of the chloroderivatives E−Te$\cdots$Cl (E = S, Se, Table 7), the S−Te and Se−Te bonds are only 0.077 Å and 0.080 Å longer than the sum of the covalent radii, making the structural features of these compounds similar to those of the fragments Se−Se$\cdots$O [358] and NC−Se$\cdots$I (see below in the last section). Finally, five structures containing the linear Te−Te−I arrangement have been reported in the literature; two of them [437] are inserted in molybdenum clusters in a fashion similar to that found for the S−S−X and Se−Se−X groups, two are arranged to form $(\text{Ph−Te−I})_4$

Table 7: Examples of the shortening of the S−Te and Se−Te bonds on passing from I to Br and to Cl derivatives.

	X	d(S–Te)	References
cis(tu)$_2$−Te−X$_2$	Cl	2.457 Å	[317]
		2.480	[377]
	Br	2.468	[377]
		2.476	[317]
	I	2.521	[317]
	X	d(Se–Te)	References
 $R_3PSe{-}\underset{\displaystyle Ph}{\underset{\vert}{Te}}{-}X^*$	Cl	2.600	[363]
	Br	2.769	[363]
	I	2.906	[439]

*R = morpholino for X = Cl and Br; R = butyl for X = I.

tetramers [435, 436] and only one, $(Mes)_2Te{-}Te(Mes){-}I$ [434], can be considered as derived from the three aligned closed-shell Mes_2Te, $MesTe^-$, and I^- species, by the removal of a couple of electrons. The analysis of the structural features of all these fragments is consistent with their description as three-body systems, the central Te atom being hypervalent.

5.2. E−X−E′ *fragments*

Table 8 collects the structural features of all the linear E−X−E′ (E, E′ = chalcogen atom; X = halogen) fragments found by searching the Cambridge Structural Database.

Systems of this type have been found with all the three halogens (Cl, Br, and I), and all the fragments have the same chalcogen (E = E′) atom at the two sides of the halogen; no mixed species (E ≠ E′) have been reported until now. Moreover, from the data in Table 8 it is interesting to note that with the exception of RIWDUW [473] which is polymeric and shows three different couples of fairly asymmetric Te−Cl bonds, all the other compounds feature the two chalcogen atoms bound to the central halogen in symmetric or only slightly asymmetric fashion, and most of the angles are very close to 180°. In all cases, the lengthening of the E−X bond with respect to the sum of the covalent radii (the mean S−I bond length calculated from the structural data of all six compounds characterized by the S−I−S group is elongated of about 17%), the shortening with respect to the van der Waals radii (the mean S−I bond length is shortened by ∼ 30%) and the linearity of the systems are consistent with the hypervalency of the central atom.

6. CHALCOGEN-DIHALIDES

Analogously to dichalcogen-halides, there are only two possibilities to build chalcogen-dihalides moieties: the chalcogen can be in the terminal (E−X−Y) or in the central (X−E−Y) position. These two arrangements correspond to the well-known CT and "T-shaped" adducts between chalcogen donors and dihalogens, respectively, and will be discussed separately.

6.1. E−X−Y *fragments*

For a more detailed discussion on this class of compounds the reader is referred to the review by Lippolis and Isaia [474]. The number of linear E−X−Y CT fragments crystallographically characterized from a search on the Cambridge Structural Database is reported in Table 9.

As one can see most of the adducts are obtained between sulphur donors (D) and diiodine, on the contrary, no compounds of this type are known with Te donors (the only reported structures featuring a Te−I−I arrangement are characterized by long I · · · I contacts). The $n\sigma(D) \rightarrow \sigma^*(XY)$ charge-transfer model accounts very well for the chemical bond in these E−X−Y systems. Scheme 2 can be easily adapted to any type of donor/acceptor couple [with the substitutions of np with $n\sigma(D)$ and $\sigma^*(X_2)$ with $\sigma^*(XY)$], bearing in mind that each couple will have a proper match of energy between the interacting orbitals. We will focus our attention on the adducts between sulfur donors and I_2, since for them it is possible to fine tune the lone pair energy of the donor atom by changing its chemical surrounding; therefore any type of adduct from very weak to extremely strong can be obtained. In the case of very weak interactions, each fragment holds its identity with a small reciprocal perturbation; the effect of such perturbation on the halogen molecule consists in the lowering to some extent of its bond order. In terms of the simplified MO diagram reported in Scheme 2, weak adducts correspond to a poor energy match between the interacting $n\sigma(D)$ and $\sigma^*(I_2)$ MO orbitals. Most of adducts between sulfur compounds and I_2 belong to the class of weak adducts. Since the stabilization of the adduct only depends on the in-phase combination of the interacting orbitals, which is bonding between the donor atom and the central iodine, and antibonding between the two iodine atoms, the two bond lengths are strictly correlated and a shortening in the D · · · I bond distance is accompanied by a lengthening in the I−I one. Without doubt, such types of adducts must be considered two-coordinate hypervalent compounds of iodine, like I_3^-. However, there is a substantial difference between an I_3^- and a D−I−I system; while in the case of I_3^- the introduction of an asymmetry, by increasing removal of one terminal iodine as I^-, generates in the limit case a strongly asymmetric $I^- \cdots I_2$ system, in the case of the charge-transfer adducts, two different asymmetric systems can be generated depending on which bond, D · · · I or I · · · I, is the weakest one. They correspond to two different charge-transfer adducts: $n\sigma(D) \rightarrow \sigma^*(I_2)$ and $n\sigma(I^-) \rightarrow \sigma^*(I{-}D)$. It is possible to pass almost continuously from a balanced situation with the two bonds having a bond order value of about 0.5 [10-I-2 "hypervalent system" for analogy to I_3^-], to the two different limit cases in which one bond assumes an increasingly ionic character. Consequently, also these limit cases featuring a strong asymmetry between the two bonds must be included among the 10-I-2 hypervalent compounds $D{-}I^+ \cdots I^-$ and $D \cdots I{-}I$.

Table 8: Structural features of all the dichalcogen-halogen (E−X−E′) fragments determined by X-ray diffraction analysis (the E−X · · · E′ fragments have not been reported).

Compound reference code	E	X	E′	d(E−X) (Å)	d(X−E′) (Å)	∠ E−X−E′(°)§	References
HAKJAE	S	I	S	2.601	2.634	175.0	[459]
IBOCUX	S	I	S	2.644	2.685	171.9	[460]
IOENCO	S	I	S	2.610	2.610	173.0	[461]
ISUREA10	S	I	S	2.629	2.629	180.0	[462]
LOPQAI	S	I	S	2.638*	2.618*	179.0*	[463]
XORVRAB	S	I	S	2.654	2.654	180.0	[464]
GIGBED	S	I	S	2.406	3.211	175.6	[465]
DIJYUQ	Se	I	Se	2.767	2.737	170.3	[466]
EZOXUM	Se	I	Se	2.765	2.765	180.0	[467]
HAKHUW	Se	I	Se	2.800	2.719	178.0	[459]
CEMFAB10	Te	I	Te	3.124	3.100	189.0	[468]
LAQZEI$	Se	Cl	Se	2.537	2.805	175.8	[469]
GANGIL**	Se	Br	Se	2.608	2.606	175.9	[470]
VIYRIE**	Se	Br	Se	2.615	2.573	176.1	[471]
MUHGUR	Se	Br	Se	3.089*	3.083*	178.8*	[472]
RIFNUP	Te	Cl	Te	2.755	2.755	180.0	[142]
ZUNJAT	Te	Cl	Te	2.857	2.829	171.4	[362]
RIWDUW‡	Te	Cl	Te	2.664*	2.988*	172.3*	[473]

§The angle values are rounded off to the first decimal. *Mean values. **The Se−Br−Se arrangement is part of the $Br_{14}Se_4^{2-}$ anion. ‡Polymeric structure. $The Se−Cl−Se arrangement is part of the $SeCl_5^-$ anion.

The scatter plot of d(S−I) versus d(I−I) relative to all the reported adducts between sulphur donors and diiodine is reported in Figure 16.

Apart for some dispersion of the data, which was also found for the other examined three-body systems, it clearly appears that the two bond lengths are strictly correlated in a wide range of values.[6] Similar correlations have been found in the case of adducts of selenium donors with diiodine (Figure 17) and sulfur donors with IBr (Figure 18), well represented in the literature. Due to the paucity of experimental data, no correlation is evident in the analogous scatter plots for the linear adducts of chalcogen donors with the other dihalogen/interhalogens molecules, including the case of d(Te−I) versus d(I · · · I). The structural features of less common linear adducts between chalcogen donors and dihalogen/interhalogens molecules are collected in Table 10.

6.2. X−E−Y *fragments*

This arrangement corresponds to the well-known "T-shaped" adducts between chalcogen-donors and dihalogens. The numbers of linear X−E−Y fragments crystallographically characterized and found by searching the Cambridge Structural Database are reported in Table 11.

While all the dihalogens/interhalogens combinations with selenium and tellurium have been reported in the literature, only few X−S−X moieties (X = Cl, Br),[7] and no X−S−Y (X ≠ Y = halogen atoms) arrangemets with sulphur as central atom are known. Indeed, sulphur donors show a preference to form linear charge-transfer type arrangement with the halogens (Table 9), absolutely unknown for the tellurium donors. Since several types of linear X−E−Y fragments are very numerous, the corresponding structural data

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[6] The points having the S−I distance higher than 3.0 Å refer to quite different systems having the diiodine molecule bridging two donor molecules.

[7] A peculiar case is represented by a byproduct of the reaction between the neutral dithiolene [Pd(Et_2timdt)$_2$] (Et_2timdt = formally monoreduced diethylimidazolidine-2,4,5-trithione) and an excess of bromine, $[(Et_2timdt)Br]_2^{+2}(Br^-)_2 \cdot (Br_2)_3$, containing two [L−S−Br]$^+$ thionebromosulfanyl cations [475]. We described this compound as ionic couples formed by the organic framework bearing a double positive charge and the two terminal S−Br bonds, and the two Br^- ions which are positioned next to the sulfur atoms to afford two very asymmetric linear Br−S · · · Br moieties. The strong asymmetry is determined by several contacts between the Br^- and Br_2 molecules. Thus, each Br^- lies in a *pseudo*-octahedral environment determined by four Br_2 molecules and two S−Br groups all acting as acceptors of the Br^- electron density. It is interesting to note that the Br_2 molecules are so firmly held in the crystal that after one year at room temperature and at air the crystals were unchanged without loss of bromine. The great stability of the Br_2 molecules within the crystal lattice can be explained in terms of donor/acceptor interaction between Br^- and six σ^* antibonding molecular orbitals of four Br_2 molecules and two S−Br terminal groups. This type of linear interaction $np(Br^-) \rightarrow \sigma^*(Br_2)$ is different from that observed in the solid Br_2 or I_2 where the halogen molecules interact each other at 90° through a $\pi^*(X_2) \rightarrow \sigma^*(X_2)$ donation (X = Br, I).

Table 9: Occurrence of linear E−X−Y CT fragments crystallographically characterized from a search of the Cambridge Structural Database (number of crystal structures in parentheses).

X−Y ↓		E ⟶	Te	Se	S
I	I		9 (9)$^{(a)}$	32 (24)$^{(b)}$	120 (85)$^{(c)}$
I	Br		—	5 (5)$^{(d)}$	13 (11)$^{(e)}$
I	Cl		—	3 (3)$^{(f)}$	4 (4)$^{(g)}$
Br	Br		3 (3)$^{(h)}$	8 (6)$^{(i)}$	9 (9)$^{(j)}$
Br	Cl		—	—	—
Cl	Cl		3 (3)$^{(k)}$	—	—

(a) Only contacts. References [476–483].
(b) References [42, 444, 466, 484–498].
(c) References [47, 69, 108, 463, 464, 474, 490, 493, 496, 499–549].
(d) References [30, 42, 148]. (e) References [25, 47, 69, 510, 550–554].
(f) References [148, 555, 556]. (g) References [510, 553–555].
(h) Only contacts. References [480, 557, 558].
(i) Only the structure of [559] is a CT adduct.
References [50, 559–561].
(j) Only two structures are of the CT type.
References [45, 63, 475, 532, 562–564].
(k) Only contacts. References [360, 483, 565].

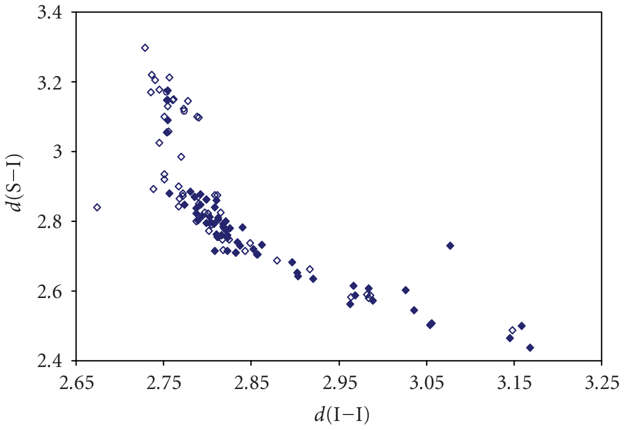

Figure 16: Scatter plot of $d(\text{S}-\text{I})$ versus $d(\text{I}-\text{I})$ for linear (angle > 165°) S−I−I fragments from a search of the CSD. The symbol (◆) refers to the 67 S−I−I fragments (50 structures) featuring bond distances ranging from Σr_{cov} to $(\Sigma r_{\text{VdW}} - 0.6)$; the symbol (◇) refers to the 50 S···I−I fragments (38 structures) featuring S···I contact distances shorter than $(\Sigma r_{\text{VdW}} - 0.3)$; the symbol (∘) refers to the 3 S−I···I fragments (3 structures) featuring I···I contact distances shorter than $(\Sigma r_{\text{VdW}} - 0.3)$. The mean bond lengthening within S−I−I fragments is 12.2% (19.2% on S···I−I, and 14.1% on S−I···I fragments, resp.) with respect to the sum of the covalent radii.

are given as scatter plots of the two X−E and E−Y bond lengths (Figures 19, 20, 21, 22, and 23).

As one can see, there is a high dispersion of points in the scatter plots; however, in all the analyzed three-body systems the two bond lengths can be considered correlated and both strongly asymmetric and symmetric fragments can be found.

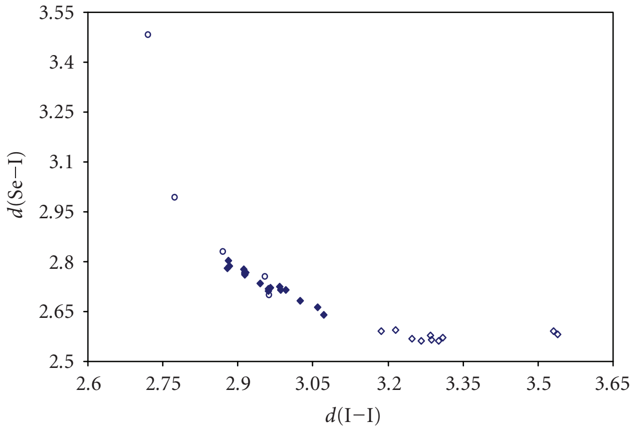

Figure 17: Scatter plot of $d(\text{Se}-\text{I})$ versus $d(\text{I}-\text{I})$ for linear (angle > 165°) Se−I−I fragments from a search of the CSD. The symbol (◆) refers to the 16 Se−I−I fragments (12 structures) featuring bond distances ranging from Σr_{cov} to $(\Sigma r_{\text{VdW}} - 0.6)$; the symbol (◇) refers to the 10 Se···I−I fragments (5 structures) featuring Se···I contact distances shorter than $(\Sigma r_{\text{VdW}} - 0.3)$; the symbol (∘) refers to the 6 Se−I···I fragments (6 structures) featuring I···I contact distances shorter than $(\Sigma r_{\text{VdW}} - 0.3)$. The mean bond lengthening within Se−I−I fragments is 10.5% (12.4% on Se···I−I, and 14.4% on Se−I···I fragments, resp.) with respect to the sum of the covalent radii.

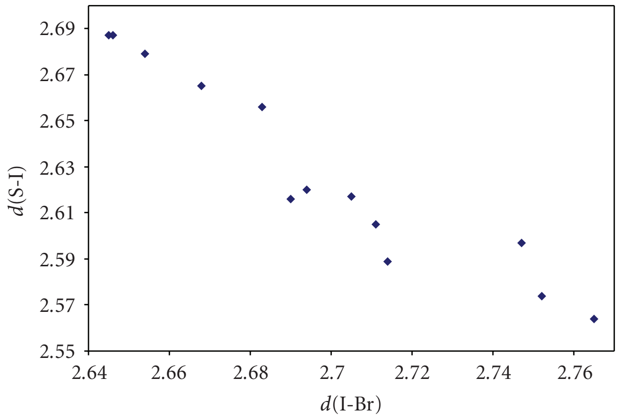

Figure 18: Scatter plot of $d(\text{I}-\text{Br})$ versus $d(\text{S}-\text{I})$ for the 13 linear (angle > 165°) S−I−Br fragments (11 structures) featuring bond distances ranging from Σr_{cov} to $(\Sigma r_{\text{VdW}} - 0.6)$ from a search of the CSD. The mean bond lengthening within S−I−Br fragments is 10.5% with respect to the sum of the covalent radii.

In Table 12, the structural features of less common X−E−Y fragments are reported; there are six examples of hypervalent chalcogen atoms bonded to two different halogen atoms, and, as already said, none of them features a central sulphur atom. It is interesting to note that in such systems the bond between the chalcogen and the lighter halogen is much more elongated with respect to the sum of the covalent radii (more ionic bond) than that involving the heavier halogen. In SUSMIC and in IDAZUI, Se−Cl and Se−Br are even longer than Se−Br and Se−I, respectively.

Table 10: Structural features of less common E−X−Y linear chalcogendihalides of the CT type, characterized by X-ray diffraction analysis.

Compound reference code	E	X	Y	E−X (Å)	X−Y (Å)	∠ E−X−Y(°)(a)	References
HAMCII	S	I	Cl	2.534	2.761	176.4	[510]
LIFXIH	S	I	Cl	2.556	2.604	179.9	[555]
NAHQIX	S	I	Cl	2.575	2.558	176.1	[553]
SIBJOC	S	I	Cl	2.641	2.586	174.9	[554]
RORNIV(b)	S	Br	Br	2.299	2.717	175.0	[562]
RORNIV01(b)	S	Br	Br	2.328	2.705	176.0	[63]
IRABEI(c)	Se	Br	Br	2.645	2.358	174.2	[559]
LIGFIQ	Se	I	Cl	2.625	2.690	178.9	[555]
LIGFIQ01	Se	I	Cl	2.618	2.690	178.7	[148]
OXSEIC	Se	I	Cl	2.630	2.731	175.8	[556]
NOWLOA	Se	I	Br	2.808	2.641	177.3	[30]
NOWLUG	Se	I	Br	2.664	2.797	175.8	[30]
WIPPAM	Se	I	Br	2.636	2.813	177.1	[148]
YEYFIR	Se	I	Br	2.689	2.908	176.9	[42]

(a)The angle values are rounded off to the first decimal. (b)Polymorphs. (c)This is the unique example of CT type adduct between a selenium donor with bromine: the formation of a Br−Se−Br group determines very favorable electronic and steric effects to prevent the formation of the same arrangement on the second selenium atom and to promote the CT type adduct. It must be noted that the Se · · · Br interaction is enough weak to determine a lengthening of the Br−Br bond of only 0.078 Å.

Table 11: Occurrence of linear X−E−Y fragments crystallographically characterized from a search of the Cambridge Structural Database.

X ↓	Y ↓	E →	Te	Se	S
Cl	Cl		194(a)	53(b)	4(c)
Cl	Br		1(d)	2(e)	—
Cl	I		1(f)	—	—
Br	Br		84(g)	63(h)	3(i)
Br	I		1(j)	1(k)	—
I	I		71(l)	4(m)	—

(a)References [142, 169, 173, 180, 181, 184, 188–190, 203, 206, 217, 219, 233, 240, 360–362, 368, 373, 374, 376, 381, 473, 480, 481, 565–699]. (b)References [260, 325, 381, 469, 495, 561, 700–729]. (c)References [730–732]. (d)Reference [733]. (e)Reference [723]. (f)Reference [580]. (g)References [61, 183, 217, 220, 232, 233, 240, 318, 373, 411, 417, 480, 481, 558, 569, 574, 607, 615, 625, 627, 635, 651, 669, 675, 688, 691, 734–777].

(h)References [50, 76, 96, 419, 421, 470, 471, 495, 559–561, 700, 718, 723, 724, 727, 752, 778–799]. (i)References [47, 788, 800]. (j)Reference [801]. (k)Reference [802].

(l)References [183, 207, 220, 233, 240, 317, 373, 435, 442, 468, 476–483, 593, 635, 672, 680, 753, 770, 801, 803–829].

(m)References [486, 498, 830].

7. CHALCOGEN · XCN (X = HALOGEN) ADDUCTS

As reported before, we wish also to consider in this discussion "T-shaped" adducts obtained from the reaction between chalcogen donors and *pseudo*-halogens X−CN (X = Cl, Br and I). Some compounds characterized by X-ray diffraction analysis and featuring X−E−CN moieties (X = halogen, E = chalcogen) are collected in Table 13.

The compound CYMIMB, reported by Arduengo and Burgess [800], has been included in the table for its strict similarity with EZUZII, reported by us [832]. Both compounds have a "T-shaped" arrangement around the chalcogen atom and are characterized by very different E−X and E−CN bond lengths; the chalcogen-carbon bond is only sligthly elongated with respect to the sum of covalent radii (bond order close to 1) and the chalcogen-halogen bond is close to be a completely ionic bond. These compounds closely resemble many asymmetric systems above decribed and in particular the pincer-type molecule bearing the O · · · Se−Se group (Table 4). The closeness of the chalcogen−CN and the Se−Se bond distances to the corresponding single bonds, respectively, and the long chalcogen-halogen and selenium-oxygen distances, strongly support the analogy between these two classes of compounds. According to the 3c-4e model, the different energy levels of the three combined *p* orbitals (there is a good overlap between the orbitals from E and C due to a good match of their energies) produce a bonding MO having a small contribution of the *p* orbital of the halogen, which vice versa mainly contributes to the nonbonding orbital, thus carrying most of the negative charge. In terms of the charge-transfer model, all the compounds of this type can be properly described as originated by a very weak donation from one halide orbital to the E−CN antibonding orbital (e.g., $n\sigma(I^-) \rightarrow \sigma^*(E{-}CN)$); the weak interaction has the consequence of a small lengthening in the E−CN bond distance, exactly as verified in numerous adducts between weak S donors and diiodine.

Table 12: Structural features of the less common X−E−Y linear chalcogendihalides characterized by X-ray diffraction analysis.

Compound reference code	X	E	Y	d(X−E) (Å)	d(E−Y) (Å)	∠ X−E−Y(°)[a]	References
CFMBXT	Cl	S	Cl	2.126	2.552	167.6	[730]
CLPHSC10	Cl	S	Cl	2.256	2.322	174.9	[731]
TOSXII	Cl	S	Cl	2.341	2.384	166.3	[732]
TOSXOO	Cl	S	Cl	2.295	2.365	175.9	[732]
BIMMAL	Br	S	Br	2.437	2.495	171.6	[47]
MIMZDB	Br	S	Br	2.451	2.538	176.9	[800]
OBUQEH	Br	S	Br	2.493	2.493	179.4	[788]
SUSMIC	Cl	Se	Br	2.802[b]	2.412[b]	173.3[b]	[723]
SUSNAV	Cl	Se	Br	2.466	2.571	176.2	[793]
IDAZUI[c]	Br	Se	I	2.831[b]	2.618[b]	174.6[b]	[802]
GEPPUM	I	Se	I	2.756	2.850	176.3	[486]
HELDUX	I	Se	I	2.768	2.854	175.4	[830]
ZOBDID	I	Se	I	2.738	2.886	178.6	[498]
ZOBDUP	I	Se	I	2.743	2.900	177.5	[498]
XAGVIK	Cl	Te	Br	2.659	2.577	169.9	[733]
CEFREX	Br	Te	I	2.868	2.903	177.9	[580]

[a] The angle values are rounded off to the first decimal. [b] Mean values. [c] This compound is the unique example of Se-hypervalent compound with IBr. Note that the mean value of the Se−I bond length is shorter than the Se−Br one.

Table 13: Structural features of all the T-shaped compounds containing the X · · · E−CN fragment (E = chalcogen; X = halogen) from a search of the Cambridge Structural Database.

Compound reference code	X	E	X · · · E (Å)	E−CN (Å)	∠ X · · · E−Y(°)	References
BOJPUL	Cl	Te	2.924	2.140	167.9	[831]
BOJRAT	Br	Te	3.100	2.131	167.6	[831]
BOJREX	I	Te	3.299	2.143	170.9	[831]
CYMIMB[a]	Br	S	3.588[a]	1.757	159.8	[800]
EZUZII	I	Se	3.300	1.885	174.8	[832]

[a] This compound has not been found searching the Cambridge Structural Database, but has been included in the table for the strict similarity with EZUZII. In CYMIMB, the shorter S · · · Br distance (3.270 Å) is that of the bromide in *trans* position with respect to the pentaatomic ring of the donor and not to the CN group.

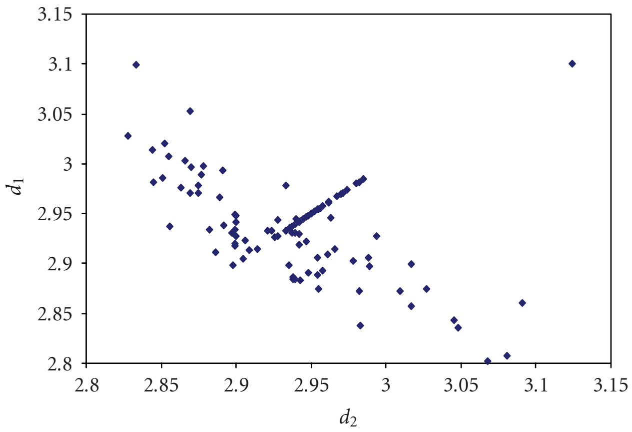

Figure 19: Scatter plot of d_1 versus d_2 of the 113 linear (angle > 165°) I−Te−I fragments (71 structures) featuring bond distances ranging from Σr_{cov} to (Σr_{VdW} − 0.6) from a search of the CSD. The mean bond lengthening within I−Te−I fragments is 9.3% with respect to the sum of the covalent radii.

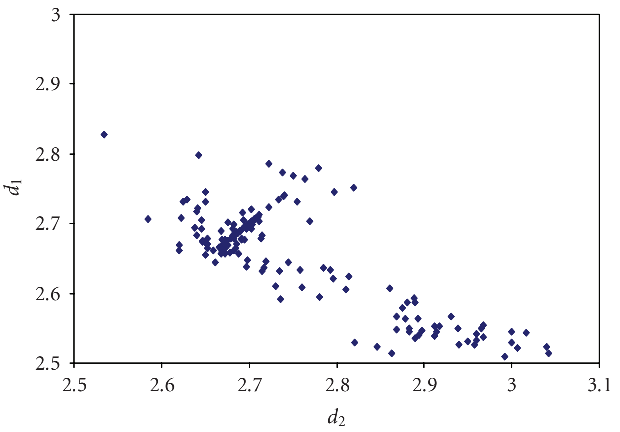

Figure 20: Scatter plot of d_1 versus d_2 of the 170 linear (angle > 165°) Br−Te−Br fragments (84 structures) featuring bond distances ranging from Σr_{cov} to (Σr_{VdW} − 0.6) from a search of the CSD. The mean bond lengthening within Br−Te−Br fragments is 8.1% with respect to the sum of the covalent radii.

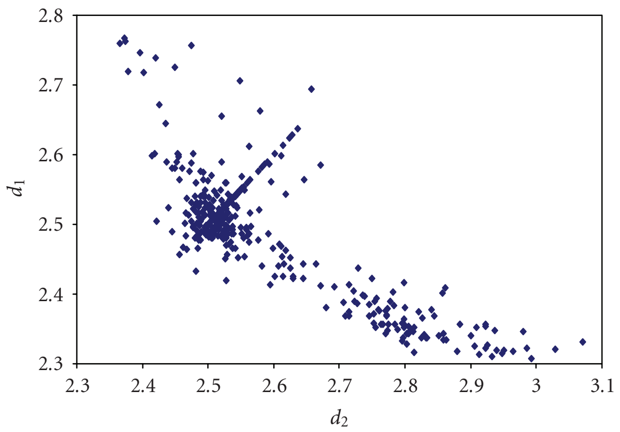

FIGURE 21: Scatter plot of d_1 versus d_2 for the 405 linear (angle > 165°) Cl−Te−Cl fragments (174 structures) featuring bond distances ranging from Σr_{cov} to $(\Sigma r_{VdW} - 0.6)$ from a search of the CSD. The mean bond lengthening within Cl−Te−Cl fragments is 8.0% with respect to the sum of the covalent radii.

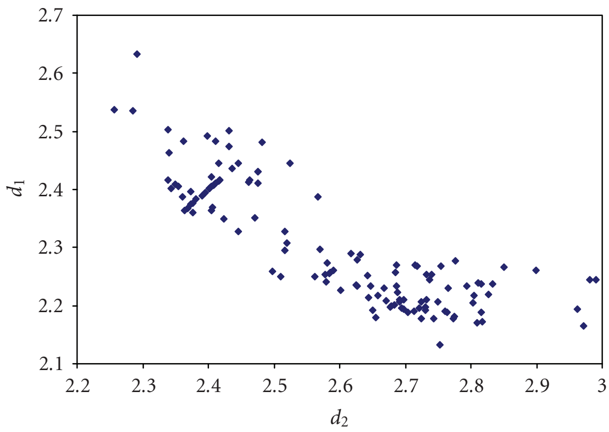

FIGURE 23: Scatter plot of d_1 versus d_2 for the 130 linear (angle > 165°) Cl−Se−Cl fragments (51 structures) featuring bond distances ranging from Σr_{cov} to $(\Sigma r_{VdW} - 0.6)$ from a search of the CSD. The mean bond lengthening within Cl−Se−Cl fragments is 13.8% with respect to the sum of the covalent radii.

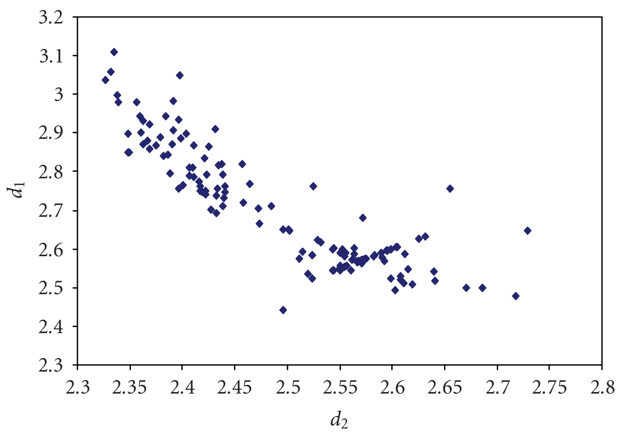

FIGURE 22: Scatter plot of d_1 versus d_2 for the 141 linear (angle > 165°) Br−Se−Br fragments (63 structures) featuring bond distances ranging from Σr_{cov} to $(\Sigma r_{VdW} - 0.6)$ from a search of the CSD. The mean bond lengthening within Br−Se−Br fragments is 12.9% with respect to the sum of the covalent radii.

8. CONCLUSION

On the basis of this overview on the structural features of linear three-body systems, involving 16th and 17th group elements, the following conclusions can be drawn.

(i) The Rundle-Pimentel model for electron-rich 3-centre 4-electron systems and the charge-transfer model represent two different approaches able to account for the structural features of these linear three-body systems.

(ii) The Rundle-Pimentel model can be adapted to any set of three aligned atoms, positioning the combining orbitals at the appropriate levels of energy.

(iii) Since three aligned closed-shell atoms can find stabilization only if two electrons are removed from the system, ideally any type of sequence of atoms could be obtained.

(vi) The variability of starting molecules is reflected in the great variety of obtainable structural archetypes. Since a starting molecule can be also a species containing a hypervalent atom, its alignment with other closed-shell species produces molecules in which two or three orthogonal 3c-4e systems are simultaneous present. This is for example the case of anions such as Ph−$SeBr_4^-$ or $SeBr_6^{2-}$.

(v) The Rundle-Pimentel model very well accounts for the 0.5 bond order in symmetric three-body systems since only the lowest MO contributes to the bond formation. In addition, the Rundle-Pimentel model elegantly explains why the two terminal atoms carry more negative charge (or less positive charge for positively charged systems) even in the cases of three identical atoms [such as I_3^- or $E(R_2)-E(R_2)-E(R_2)^{2+}$ dications].

(vi) In these three-body systems, the energy match between the *p* orbital of the central atom and those of the terminal ones influences the polarization of the formed bonds. This is very important for systems having different terminal atoms: each *p* orbital will contribute differently to the three molecular orbitals with the consequence of an increased unbalance of the two bonds as the electronegativity difference between the involved elements increases. In such cases, the bond orders of the two bonds diverge from the value of 0.5, one approaching the value of 1 and the other that of 0.

(vii) The strict analogy among all these systems, including the strongly asymmetric ones as the "T-shaped" adduct between the N, N′-dimethylimidazoline-2-selone and ICN, supports the hypervalent nature of the selenium atom in this compound in spite of the fact that the bond orders of the two bonds are very different.

(viii) The charge-transfer model explains very well all the very asymmetric systems since this model corresponds to the interaction of two stable fragments (as a dihalogen molecule with a halide, or as chalcogen donor with a dichalcogen dication).

(ix) The energy match between the interacting orbitals of the two fragments (such as a hybrid orbital of the donor and the σ^* antibonding molecular orbital of the acceptor) determines the entity of the interaction.

(x) In the CT model, the bond order of 0.5 for the two bonds is reached when the interacting orbitals are at the same level of energy. This corresponds to the introduction of 1 electron on the σ^* MO of the acceptor, with the consequent reduction of the bond order from 1 to 0.5.

(xi) An aspect to be emphasized is the fact that in all the structures of these families of compounds, including the very asymmetric systems, the three-body system is always linear, with angles generally larger than 170°. The directionality of the bond is maintained also in presence of strongly unbalanced bonds indicating a valuable contribution of covalence, due to the $n\sigma_{\text{donor}} \rightarrow \sigma^*_{\text{acceptor}}$ charge-transfer interaction and supporting the hypervalent character of the central chalcogen atom, independently on the entity of the asymmetry.

(xii) Finally, it is interesting to observe that with only few exception, the systems having different terminal atoms (see, e.g., trihalides $X{-}Z{-}Y$ with $X \neq Y$ or trichalcogenides $E{-}E'{-}E''$ with $E \neq E''$) are less common than the symmetric ones.

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